EFFECT OF SOME ANTIOXIDANTS ON THE WEIGHT CHANGE OF LINOLEIC AND LINOLENIC ACIDS AND THEIR DERIVATIVES IN THE COURSE OF AUTOXIDATION

YOSHIYUKI TOYAMA and YOSHIO HIRABAYASHI*

Department of Applied Chemistry

(Received May 30, 1954)

In the preceding paper,[1] the weight change of the sample material in the course of the indoor autoxidation without exposure to sun light and the outdoor autoxidation under exposure to sun light was studied by using linoleic acid, methyl and ethyl linoleate, linoleyl alcohol and its acetate, methyl heptadecadienyl ketone (10,13-nonadecadien-2-one) and the corresponding compounds of linolenic series as the sample materials. The present paper deals with the effect of some antioxidants on the weight change of these compounds in the course of autoxidation. The experiments described in this paper were performed in the same way as the indoor experiments described in the preceding paper using the same samples. Antioxidants used in these experiments are pyrogallol, pyrocatechol, hydroquinone, resorcinol, thymol, α- and β-naphthols, *p*-toluidine, β-naphtylamine and diphenylamine. They were added to the sample in an amount of 0.2%.

Experimental Results and Discussion

The results of experiments are shown in Figs. 1–12. Although the effectiveness of each antioxidant is somewhat different for each sample under autoxidation, pyrogallol, pyrocatechol and α-naphthol are most effective for all samples. Hydroquinone stands next to these three antioxidants in its effectiveness. This is consistent, on the whole, with the results previously reported by Ono and Toyama[2] for the autoxidation of highly unsaturated acids separated from sardine oil that the antioxidant activity is in the order of pyrogallol > α-naphthol > pyrocatechol > hydroquinone. It should be noted, however, that while the highly unsaturated acids of sardine oil added with diphenylamine had a remarkably small rate of weight increase over a certain range of autoxidation showing a peculiar weight increase curve, this characteristic property of diphenylamine does not appear in the present experiments with the compounds of linoleic and linolenic series. Resorcinol is in most cases inferior to hydroquinone in its antioxidant effect. Thymol has the least activity among the phenols examined. Comparing α- and β-naphthols, the former has a greater activity. *p*-Toluidine has the least activity among the anti-

* Present address: Department of Industrial Chemistry, Faculty of Engineering, Gifu University.

oxidants examined, and is almost ineffective for methyl linoleate, linolenyl acetate and methyl heptadecatrienyl ketone (10, 13, 16-nonadecatrien-2-one).

The curves in Figs. 1–12 are marked as follows:

Numerical numbers 1–6: the samples without antioxidant; *Pg*: pyrogallol; *Th*: thymol; *Pc*: pyrocatechol; *H*: hydroquinone: *R*: resorcinol; *α*: *α*-naphthol; *β*: *β*-naphthol; *To*: *p*-toluidine; *βN*: *β*-naphthylamine; *D*: diphenylamine.

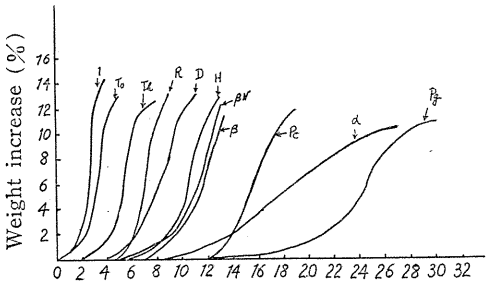

FIG. 1. Linoleic acid

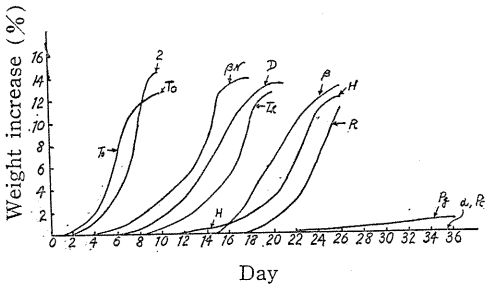

FIG. 2. Methyl linoleate

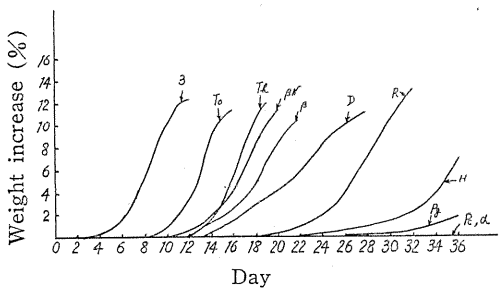

FIG. 3. Ethyl linoleate

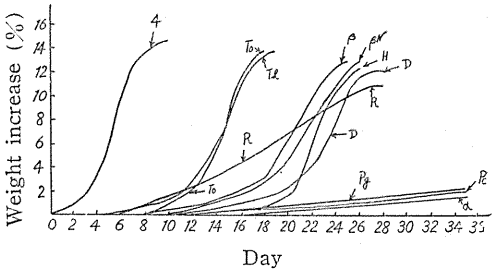

FIG. 4. Linoleyl alcohol

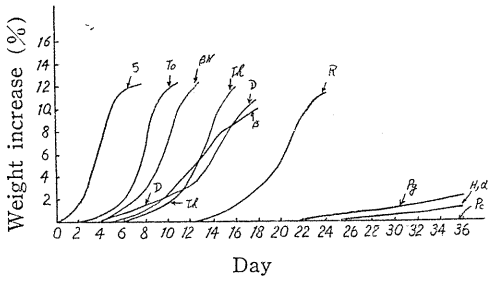

FIG. 5. Linoleyl acetate

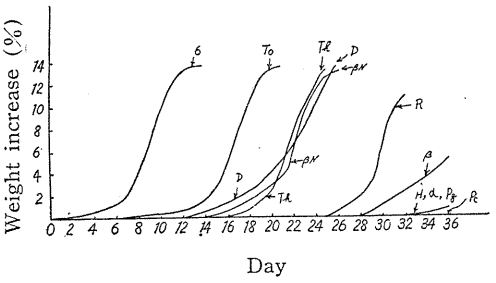

FIG. 6. 10, 13-Nonadecadien-2-one

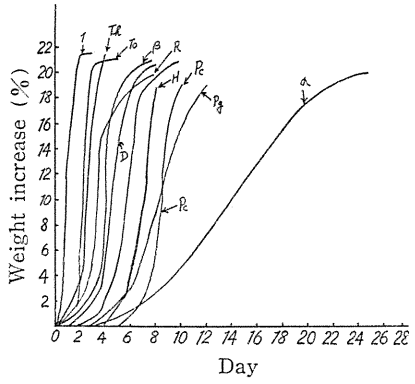

FIG. 7. Linolenic acid

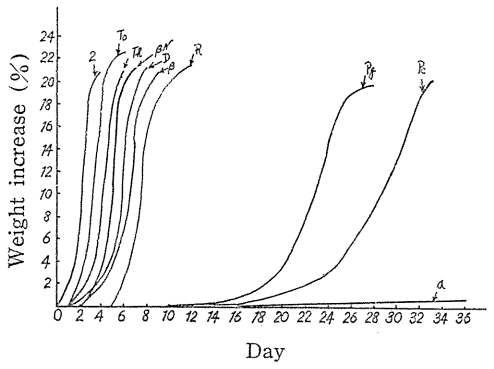

FIG. 8. Methyl linolenate

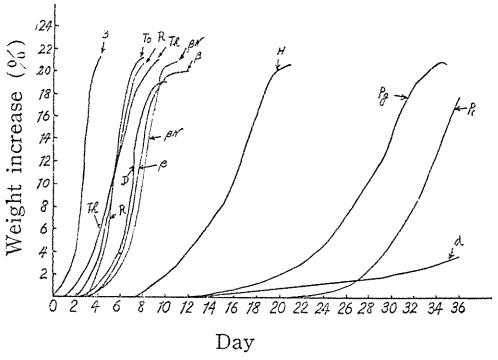

FIG. 9. Ethyl lonolenate

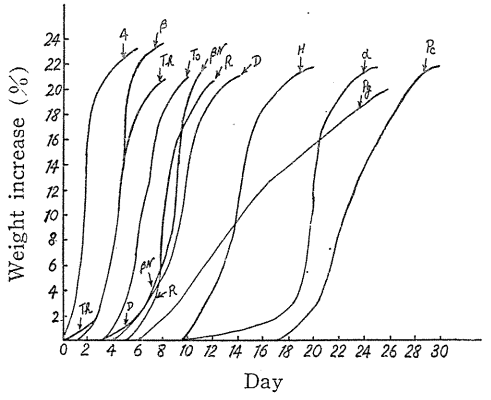

FIG. 10. Linolenyl alcohol

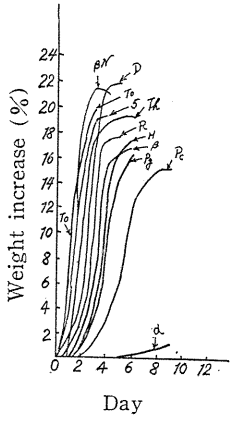

FIG. 11. Linolenyl acetate

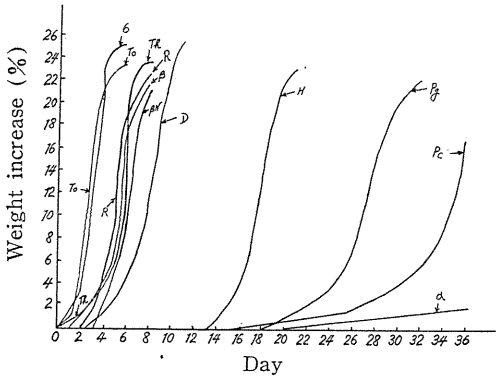

FIG. 12. 10, 13, 16-Nonadecatrien-2-one

Summary

Using pyrogallol, pyrocatechol, hydroquinone, resorcinol, thymol, α- and β-naphthols, *p*-toluidine, β-naphthylamine and diphenylamine as antioxidants, their effect upon the weight change of linoleic acid and its methyl and ethyl esters, linoleyl alcohol and its acetate, 10,13-nonadecadien-2-one, and the corresponding compounds of linolenic series in the course of autoxidation was studied with the following results. Among these antioxidants, pyrogallol, pyrocatechol and α-naphthol are most effective for all compounds. Hydroquinone stands next to these three antioxidants in its effectiveness. Resorcinol is inferior to hydroquinone in most cases. Thymol has the least activity among the phenols. *p*-Toluidine is least effective among the antioxidants examined.

References

1) Y. Toyama and Y. Hirabayashi: *Memoirs Faculty of Engineering, Nagoya Univ.* **6**, 53 (1954).
2) F. Ono and Y. Toyama: *J. Chem. Soc. Japan* **65**, 611 (1944).