Occurrence of Orthophenylphenol During Manufacture of Lemon Marmalade

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(Received for publication July 17, 1978)

ABSTRACT

Orthophenylphenol (OPP) and diphenyl (DP) are effective preservatives to prevent putrefaction of citrus fruits and vegetables caused by Oidium species, and their use on various kinds of foods is approved in the U.S.A. and in EC countries with limitation. Contrary to this, in Japan since 1971 only diphenyl is allowed to be used as a preservative for grapefruit, lemon and oranges with a maximal residual level of 70 ppm. Use of OPP and other preservatives was strictly prohibited. In April 1975 quantities of lemon and other citrus fruits had to be destroyed because they contained detectable amount of OPP (1). It was quite recent (April 30, 1977) that OPP as well as its sodium salt were added to the Japanese list of acceptable chemically synthesized food additives (2). Its use, however, is restricted to citrus fruits with a maximal permitted residual level of 10 ppm as OPP.

It must be taken into account, however, that, besides being consumed raw, citrus fruits are often used as one of the raw materials in the manufacture of marmalade. At present, OPP is not allowed to be used in the manufacture of marmalade or jams. Accordingly, it was undertaken by us to ascertain the occurrence of OPP in marmalade when it was made from lemons containing permissible level of OPP.

MATERIALS AND METHODS

Preparation of lemon marmalade

Marmalade was prepared by an ordinary method in the following way. Lemons imported from the U.S.A. were cut into quarters and peeled. Peels were cut into pieces, in some instances boiled for 30 min in 1% NaCl solution to remove bitter substances, and finally partially dehydrated to obtain peel pieces for marmalade preparation. Pulps were crushed and divided into endocarps and edible portions; the former was mixed with twice as much, by weight, of 0.25% citric acid solution being followed by boiling for 30 min to obtain crude pectin while the latter was boiled for 3 min to obtain the jelly base. Crude pectin and jelly base were mixed in the ratio of 4:6; addition of peel pieces to pectin jelly mixture was arranged to obtain six kinds of final products.

Analysis of OPP

OPP contents in samples were determined gas chromatographically by use of the Standard Methods for Hygienic Chemists (3). A Yanaco G-80 gas chromatograph (Yanagimoto Mfg. Co. Ltd.) with flame ionization detector attached, being fitted with glass tubing of 1.5 m in length packed with 3% SE-30 on Chromosorb W 60/80, was used. Under this condition, recovery of OPP at the level of 1 ppm added to sample No. 1 was found to be 97.4%.

RESULTS AND DISCUSSION

OPP content in lemons used as raw material was determined, on a whole basis, to be 2.3 ppm. The values obtained on samples are summarized in Table 1. It was confirmed that 57% of OPP was lost during removal of bitter substances.

The ratio of OPP content of sample No. 3 to No. 6 was 0.45 which was nearly the same as that of sample No. 2 to No. 1. Since the composition of raw materials was the same among samples No. 1 and 2 and No. 3 and 6, it is suggested that OPP content in sample No. 6 was little affected by dipping the peels in water. Furthermore, a comparison of OPP contents was done by changing the...
TABLE 1. OPP contents in prepared jams.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Removal of bitter substances</th>
<th>Ratio of Pectin, Jelly base and Peel</th>
<th>OPP content (ppm)</th>
<th>Ratio to No. 2</th>
<th>Ratio to No. 1</th>
<th>Ratio to No. 6</th>
</tr>
</thead>
<tbody>
<tr>
<td>No. 1</td>
<td>Not carried out</td>
<td>4:6:1.0</td>
<td>0.90</td>
<td>--</td>
<td>1</td>
<td>--</td>
</tr>
<tr>
<td>No. 2</td>
<td>Carried out</td>
<td>4:6:1.0</td>
<td>0.39</td>
<td>1</td>
<td>0.43</td>
<td>1</td>
</tr>
<tr>
<td>No. 3</td>
<td>&quot;</td>
<td>4:6:1.5</td>
<td>0.54</td>
<td>1.38</td>
<td>--</td>
<td>0.45</td>
</tr>
<tr>
<td>No. 4</td>
<td>&quot;</td>
<td>4:6:2.0</td>
<td>0.82</td>
<td>2.10</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>No. 5</td>
<td>&quot;</td>
<td>4:6:3.0</td>
<td>1.13</td>
<td>2.90</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>No. 6</td>
<td>&quot;</td>
<td>4:6:1.5</td>
<td>1.21</td>
<td>--</td>
<td>--</td>
<td>1</td>
</tr>
</tbody>
</table>

*Pc stands for pectin, J for jelly base and P for peel pieces, respectively.*

**REFERENCES**


amount of peel pieces in samples No. 2-5. From the results it became clear that change in OPP content was proportional to the increase in the combined ratio of peel pieces, indicating that most of OPP contained in the raw material was in the peel fraction.

Only 17% of the initial amount of OPP remained in the finished product, lemon marmalade (cf. Sample No. 2) prepared by an ordinary procedure from lemon which contained 2.3 ppm of OPP.

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