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# QUALITY OF CLOVE BUD AND CLOVE BUD OIL

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*Keywords*: Clove bud, Clove bud oil, *Eugenia caryophyllus*, Eugenol.

**RINGKASAN**

Dua jenis bunga cengkih yang diperolehi dari pasar dan kawasan penanaman tempatan telah dikaji mutu fizik, kimia dan minyaknya. Bunga cengkih dari ladang tempatan didapati mempunyai mutu fizik yang lebih tinggi berbanding dengan jenis dari pasar walaupun sifat-sifat kimianya hampir sama. Kedua-dua jenis menghasilkan minyak sebanyak 15%–17% dengan kandungan jumlah fenol di antara 79% hingga 94 peratus. Ciri-ciri fizik minyak didapati hampir sama atau rendah sedikit daripada standard ISO.

## INTRODUCTION

Cloves are the dried flower buds of *Eugenia caryophyllus* (Spreng.) or also known as *Caryophyllus aromaticus* L., an evergreen tree belonging to the Myrtaceae family. The tree is believed to be indigenous to the Moluccas archipelago, especially the Amboyna Island (ANON., 1963; PARRY, 1969a), but is now widely cultivated in Tanzania (Zanzibar and Pemba), Madagascar, India, Sri Lanka, West Malaysia (especially the Penang Island) and several other places.

Clove has been cultivated in Malaysia for over 180 years. It was first introduced from the Moluccas archipelago in 1800 by Smith, a botanist, who brought about 15 000 young clove plants to Penang (GUENTHER, 1972b). The industry reached its peak in 1860 with more than 5 800 hectares under clove cultivation. However, it suffered a serious setback when rubber became more important. By 1970, the area under clove cultivation was slightly more than 100 hectares, with an annual production of about 45 tonnes. Since then, the area for total sole crop equivalent of clove has increased to 410 hectares in 1982 (ANON., 1983).

Although the bulk of the world supply of clove comes from Zanzibar and Madagascar, Penang and Amboyna cloves are said to command higher prices. Local cloves are larger, more plump, bright reddish-brown and more fragrant than other varieties. The Amboyna cloves though similar are smaller while Zanzibar and Madagascar cloves are much smaller, shrunken, darker and less fragrant (ANON., 1963). This study was carried out to compare the physical quality of clove buds (those available in the market and those obtained directly from the farm) and the physico-chemical characteristics of clove bud oils with the standards of the International Standard Organization (ISO) and the oils from other countries. Earlier studies by MOESLIM and ZAINAB (1974) dealt mainly with oil isolation techniques of market samples of clove buds. Very little emphasis was given to the quality of bud and oil while the physical quality was not mentioned at all.

## MATERIALS AND METHODS

### Clove Bud Samples

Two types of clove bud samples were used. Type I comprised three samples of dried clove buds of 500 g each, bought from three different retail outlets in the vicinity of Serdang and Kajang. Type II was a representative: one kilogram of matured, dried clove buds randomly sampled from the Department of Agriculture, Hutan Melintang, Perak. It was equally divided

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into three portions. All samples were then divided into two equal lots. One lot from each sample was comminuted to fine powder by using a coffee grinder.

**Physical Examination**

The thoroughly mixed uncomminuted buds were examined manually to determine the percentages of whole buds (buds with intact heads), buds without heads, broken buds (with or without heads) and foreign matter.

**Chemical Analysis of the Bud**

General chemical analyses for non-volatile ether extract, total ash, acid-insoluble ash, volatile oil, crude fibre and moisture content were carried out on the clove buds according to the Standard A.O.A.C. methods 1975. Moisture content was determined by the distillation method using toluene as solvent.

**Extraction of the Oil**

The oil was extracted by direct water distillation using apparatus as shown in *Figure 1*. A 100-g sample was placed in a one-litre round-bottommed flask which was half-filled with water.

The sample was heated using an electric heating mantle. Steam and liberated oil condensed upon passing through the condenser. Oil together with water were trapped at the graduated column from which excess water of condensation flowed back to the flask via the back-flow column. Distillation lasted for about four to six hours for complete oil extraction. Oil collected in graduated column was then drained out through a stopcock, collected and dried using anhydrous sodium sulphate.

**Analysis for Physico-chemical Properties of the Oil**

The physical characteristics of the oil analysed were colour, refractive index (RI),

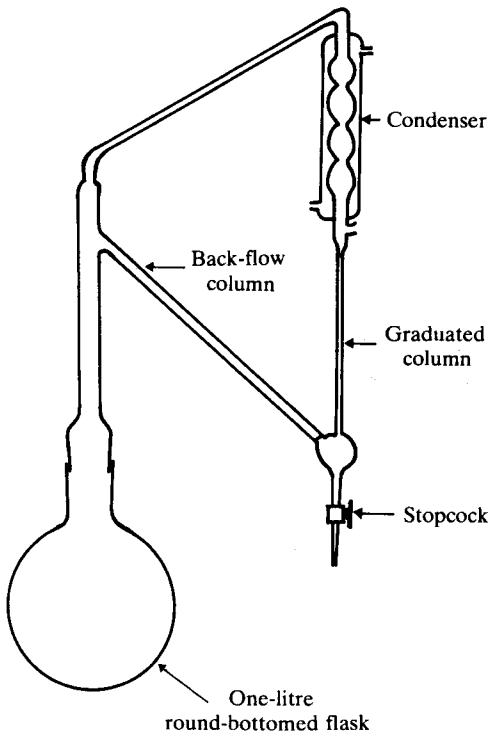


*Figure 1. Distillation unit for extraction of clove bud oil.*

specific gravity (SG) and optical rotation (OR). Colour was determined by using a Lovibond tintometer. An Abbe refractometer and a 10-ml picnometer were respectively used to determine the RI and SG at 25° Celsius. Optical rotation was measured using a polarimeter equipped with a 1-dm tube.

Phenol content of the oil, in terms of total free eugenol, was determined according to the methods described by GUENTHER (1972a). In this method, 10 ml of oil was introduced into a 150-ml Cassia flask and 75 ml of 1.0 *N* potassium hydroxide was added. The mixture was shaken vigorously for five minutes to ensure thorough extraction of phenol by the alkaline solution. It was then heated for ten minutes using a steam bath, with occasional shaking to ensure complete saponification. Immediately after removing the flask from the steam bath, a further quantity of alkali was added to drive unreacted oil into the neck of the flask. This

addition was necessary while the contents were still hot as the non-phenolic portion may partially solidify when cooled. The quantity of the unreacted oil was measured directly from the readings on the graduated neck of the Cassia flask. The phenol content, expressed as a v/v percentage, was calculated using the following formula:

% phenol (as free eugenol)
= 100 (10 − amount of unreacted oil in millilitre)

## RESULTS AND DISCUSSION

### Physical Examination of the Bud

Physical examination of the buds showed that type II had more whole buds and less buds without heads, broken buds and foreign matter than type I *(Table 1)*. Type I buds were darker brown than those of type II. The colour of type II was in agreement with that described in the British Pharmaceutical Codex (BPC) for Penang clove which commands high prices (ANON., 1963). In general, type II was superior to type I in physical appearance.

Table 1. Physical examination of clove buds

| Variable | Type I | Type II |
|---|---|---|
| Whole bud (%) | 66.1 | 90.6 |
| Bud without head (%) | 27.1 | 6.3 |
| Broken bud (%) | 1.2 | 0.7 |
| Foreign matter (%) | 5.6 | 2.4 |
| Colour | Dark brown | Light brown to brown |

Values are means of 3 samples.

Dried clove buds are graded according to appearance and percentage of impurities present before being exported. Exportable cloves are usually firm, flawless, having a uniform light brown colour and free from mould. In the world market, clove buds are usually referred to either as the Zanzibar grade (PARRY, 1969b; GUENTHER, 1972b) or the Malagasy grade (LAWRENCE, 1979) as are given in *Tables 2* and *3* respectively. Using these two grades, type I is equivalent to No. 3 of the Zanzibar grade or regular of the Malagasy grade. Type II is between No. 1 and 2 of the Zanzibar grade or between superior and standard of the Malagasy grade.

Table 2. Zanzibar grade

| Grade | Extraneous matter (%) | Khoker (%) | Moisture (%) |
|---|---|---|---|
| Special grade | 3 | 2 | 16 |
| Grade No. 1 | 5 | 3 | 16 |
| Grade No. 2 | 5 | 7 | 16 |
| Grade No. 3 | 5 | 20 | 16 |

Note: Extraneous matter include stems, mother of clove, foreign and inferior matters.
Khoker are those buds which under bad drying and storage conditions become darker, rather musty and finally pale and wrinkled.

The standards set by the US government do not permit more than 5% stems in a sample (PARRY, 1969b), whereas the BPC stated that an unground clove sample should contain not more than 1% foreign organic matter and not more than 5% stalks (ANON., 1963).

### Chemical Properties of Clove Bud

The general chemical properties of clove bud for types I and II, and the ISO standards are presented in *Table 4*. The values for non-volatile ether extract of both types were lower compared with that of ISO. Both types I and II had total ash contents of less than 5% which was lower

Table 3. Malagasy grade

| Grade | Moisture (%) | Vegetable impurities (%) | Foreign matter (%) | Cloves without final bud (%) |
|---|---|---|---|---|
| Prime | 16 | 2 | 0 | 2 |
| Superior | 16 | 3 | 0 | 3 |
| Standard | 16 | 5 | 0 | 10 |
| Regular | 16 | 5 | 1 | 20 |

Table 4. General chemical properties of clove bud

| Variable (%) | Type I | Type II | ISO standards* |
|---|---|---|---|
| Non-volatile ether extract | 3.43 | 3.83 | 4.0 – 10.0 |
| Total ash | 4.85 | 4.91 | 5.0 – 8.0 |
| Acid-insoluble ash | 0.37 | 0.05 | 0.1 – 0.6 |
| Volatile oil (dry wt. basis) | 15 – 16 | 15 – 17 | > 15 |
| Crude fibre | 8.56 | 9.16 | 10 – 13 |
| Moisture content | 9.0 | 11.5 – 12.0 | < 8 |

Values are means of 3 samples.
*Source : LAWRENCE (1979).

than the BPC standards (< 7%) and the ISO standards (5%–8%). The BPC standards permitted acid-insoluble ash of not more than one per cent. Both types I and II contained acid-insoluble ash which was far below these BPC standards. The volatile oil contents of both types of clove bud satisfy the ISO as well as the BPC standards of not less than 15 per cent. The crude fibre contents of types I and II were lower than the ISO standards (10%–13%). In general, types I and II were similar in most chemical properties except moisture content, the latter having a higher moisture content than the former.

## Yield of Oil

Types I and II clove bud had oil yields ranging from 15.2% to 16.9% (on a dry-weight basis) *(Table 5)*. Comminuted buds were found to give higher oil yields (16% or more) than whole buds (15.2%). This is mainly because comminuted buds have larger surface area which helps in releasing more oil. Zanzibar and Madagascar cloves yielded about 17% oil (GUENTHER, 1972b), whereas cloves sampled from the local market yielded 12.7%–14.0% oil (MOESLIM and ZAINAB, 1974).

Table 5. Yield of clove oil

| Sample | Yield (%) | |
|---|---|---|
| | v/w | w/w (dry basis) |
| Type I | | |
| Whole bud | 13.1 ± 1.3 | 15.2 ± 1.5 |
| Comminuted bud | 13.6 ± 3.1 | 16.0 ± 3.6 |
| Type II | | |
| Whole bud | 12.8 ± 0.4 | 15.2 ± 0.5 |
| Comminuted bud | 14.6 ± 1.6 | 16.9 ± 2.2 |

Values are means (with standard deviations) of 3 samples.

## Physico-chemical Properties of Oil

### Phenol content

The quality of clove oil is determined by its content of phenols, chiefly eugenol ($C_{10}H_{11}O_2$). Clove oil contains about 10% eugenyl acetate in addition to free eugenol, and both constituents contribute to the value of the oil. In this study, eugenyl acetate was saponified and reported as free eugenol. Total phenol contents of both whole and comminuted types I and II clove bud ranged from 79% to 94% *(Table 6)*. The ISO standards were set at 85%–93%, whereas BPC standards were set at 85%–90% (ANON., 1963). The phenol content of a clove oil depends on the condition of the cloves, whether whole or crushed, and also the method of distillation used (GUENTHER, 1972b). The results showed that whole clove bud produced oil with higher phenol contents (93%–94%) than oil obtained from comminuted samples (79%–88%) although the latter, because of larger exposed surface area, yielded more oil. GUENTHER (1972b) reported that steam distillation extracted more eugenol (91%–95%) from the oil than water distillation (85%–89%).

### Physical Characteristics

The SG for type I whole clove bud was within ISO standards while the rest were below the standards *(Table 6)*. The BPC standards stated that the SG of clove oil should range from 1.041 to 1.054. According to GUENTHER (1972b), oils with a high phenol content exhibit a high specific gravity. The SG of whole bud samples was found to be higher than the comminuted

Table 6. Physico-chemical properties of clove oil

| Variable | Type I | | Type II | | ISO standards* |
|---|---|---|---|---|---|
| | WB | CB | WB | CB | |
| Total phenol (%, v/v) | 93.0 | 88.0 | 93.8 | 79.0 | 85–93 |
| Specific gravity | 1.053 | 1.042 | 1.039 | 1.026 | 1.044–1.057 |
| Refractive index | 1.528 | 1.527 | 1.530 | 1.524 | 1.528–1.538 |
| Optical rotation | -4°42′ | -6°14′ | -0°50′ | -4°50′ | -1.5°–0° |
| Solubility in 70% ethanol (v/v) | – | – | – | – | 1:2 |
| Colour | | | | | |
| R | 0.2 | 0.2 | 0.2 | 0.1 | – |
| Y | 0.3 | 0.3 | 0.9 | 0.2 | – |

WB = Whole bud
CB = Comminuted bud
*Source : LAWRENCE (1979).

samples which had a lower phenol content than the former.

The OR values for all samples except whole bud type II (−0°50′) were very much lower than those of ISO standards. The RI for both types of whole bud samples was within the standards while that of comminuted bud samples was slightly lower. Solubility of oil in 70% ethanol was not tested. A study by MOESLIM and ZAINAB (1974) on market samples showed an oil solubility of 1:1.7–1.8. The ISO and BPC standards set oil solubility at 1:2. The colour of the oils as measured by a Lovibond tintometer was pale yellow with a value of R around 0.2 and Y around 0.3. It was observed that the colour darkened on storage. According to the BPC, freshly distilled clove oil should be colourless or pale yellow, darkening with age or becoming reddish brown on exposure to air. The darkening of oil on storage is probably due to the furfural content (ANON., 1963).

## CONCLUSION

The study showed that clove bud sampled from the market (type I) was inferior in physical appearance to samples obtained from the farms in Hutan Melintang, Perak (type II). The general appearance of type II samples conformed to the BPC standards. These samples were equivalent to grade No. 1 or 2 of the Zanzibar grade or between superior and standard of the Malagasy grade.

Apart from the slight difference in moisture content (9% for type I as compared with 12% for type II), other chemical properties of clove bud types I and II were similar and within the values of ISO standards.

Both types had oil yields of more than 15%, the minimum requirement stated in the BPC. Although comminuted samples yielded more oil, the phenol content of the oil was lower than in the oil from whole samples. All samples, except comminuted sample of type II, produced oil with phenol contents within the range of the ISO standards. The physical characteristics of the oils of all samples were similar or slightly below the ISO standards.

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## ABSTRACT

Two types of clove bud, the market samples and the locally grown samples, were studied for their physical, chemical and oil qualities. The locally grown samples were superior in the general physical appearance to the market samples although the chemical properties were similar. Both types had oil yield of about 15%–17%, with total phenol content ranging from 79% to 94 per cent. The physical characteristics of the oils were similar or slightly below the ISO standards.

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