Changes in Physicochemical Properties and Volatile Compounds of Roselle (Hibiscus sabdariffa L.) Calyx during Different Drying Methods

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Abstract: Fresh roselle are high in moisture and deteriorate easily, which makes drying important for extending shelf-life and increasing availability. This study investigated the influence of different drying methods (oven-drying, freeze-drying, vacuum-drying, and sun-drying) on the quality of roselle calyx expressed as physicochemical properties (moisture content, water activity, soluble solids, color), volatile compounds, and microstructure. Oven-drying and freeze-drying reduced moisture content most while vacuum-drying and sun-drying were not as efficient. All drying methods except sun-drying resulted in water activities low enough to ensure safety and quality. Vacuum-drying had no impact on color of the dry calyx and only small impact on color of water extract of calyx. Drying reduced terpenes, aldehydes, and esters but increased furans. This is expected to reduce fruity, floral, spicy, and green odors and increase caramel-like aroma. Sun-drying produced more ketones, alcohols, and esters. Scanning electron microscopy revealed that freeze-drying preserved the cell structure better, and freeze-dried samples resembled fresh samples most compared to other drying techniques. The study concludes that freeze-drying should be considered as a suitable drying method, especially with respect to preservation of structure.

Keywords: roselle (Hibiscus sabdariffa L.); drying; dynamic headspace sampling; physicochemical properties; volatile compounds; microstructure

1. Introduction

Roselle (Hibiscus sabdariffa L.) locally known as asam belanda, asam susur or asam paya in Malaysia, is a member of the Malvaceae family. It is originally native from India to Malaysia, but now it is widely distributed and cultivated in tropical and subtropical regions all around the world. Many parts of roselle including seeds, leaves, fruits, and roots are used in various foods. Among all, calyces are the most popular being used for making soft drinks, juices, teas, wines, jams, jellies, pickles, fruit leathers, yogurts, and colorants. Overseas demand for roselle products is also very encouraging [1,2]. In addition to the increasing popularity of roselle in healthcare, beverages, and cosmetics [3], roselle is a plant which can be used to improve the quality of food products. Calyces are rich in vitamin C, minerals and other antioxidants such as anthocyanin, flavonoids [4], and phenols [5], which are hypothesized to be beneficial to human health by preventing cancer and reducing chronic illnesses such as diabetes, dyslipidemias, high blood pressure, and coronary heart disease [6].

Drying is one approach that can be applied to prolong the shelf-life of fresh materials. Several commercial drying techniques are available, and every type has its own...
advantages and limitations and results in different nutritional and physicochemical characteristics of the final product. In addition, a good drying technique can enhance the quality of the product significantly [7]. Various drying techniques such as spray-drying [8], freeze-drying [9], solar thermodynamic drying [10], fixed bed drying [11], sun-drying, and oven-drying [12] have been applied to roselle. For dried roselle, the aroma and flavor are affected by the drying parameters and drying conditions [13]. Mostly, drying methods assure microbial stability, guarantee shelf-life of the product, and facilitate packaging and distribution [7,14,15]. Agudelo et al. [16] showed that for the manufacture of powered fruit, freeze-drying method is highly recommended. Furthermore, Martínez-Navarrete et al. [17] showed that freeze-drying is one of the methods that better preserves the bioactive compounds of the fruits, but, unfortunately, is very expensive. Therefore, studies of different drying methods are needed in order to attain the desired quality of the final product.

To our knowledge, no studies have been done to evaluate the influence of different drying methods on roselle calyx in terms of physicochemical properties, volatile compounds, and microstructure in comparison with the fresh roselle. Thus, the objective of this study was to determine the influence of oven-drying, freeze-drying, vacuum-drying, and sun-drying on the physicochemical properties, volatile compounds, and microstructure of roselle (*Hibiscus sabdariffa* L.). This study would be considered valuable when selecting appropriate drying methods for roselle with the aim of fulfilling the consumer demand for processed food products that are close to fresh samples and retain more of the original characteristics. Due to the high moisture content of fresh roselle, it is vital to produce a high-quality dried roselle calyx.

### 2. Results and Discussion

#### 2.1. Physicochemical Analysis

Table 1 shows that there were significant (*p* < 0.001) differences in all physicochemical properties measured. Fresh roselle calyx contained 89.4% of moisture (water activity 1.000). Sun-drying turned out to be the least efficient drying method since sun-dried samples had the highest moisture content (15.6%, water activity 0.727). This result was due to the weather because a haze phenomenon occurred in Malaysia from September until November 2015. The entire process of sun-drying was disturbed since the haze prevented the penetration of sunlight to the samples. All oven and freeze-dried samples had moisture contents below 10% (water activities 0.395–0.482). Materials with excessive moisture content will be susceptible to bacteria and molds but since water activity was 0.605 or lower in oven and freeze-dried samples, they were considered safe for general storage [18,19].

Extracts based on freeze-dried roselle had the highest value of total soluble solids (TSS) (Table 1). A higher value of TSS may be attributed to longer drying time and possibly pectic enzyme activity [20] which may increase the amount of soluble solids. Other factors could be better structure of the dried product due to higher porosity at lower shrinkage, which enhances the grinding process and thus extraction.

Color is an important quality attribute in roselle; in fact, roselle is used as food coloring due to the high content of anthocyanin. In this study, the effects of different drying methods on the color of dried roselle calyx were measured in ground calyces and in a liquid extract (‘filtrate’). Preferred colors are those closest to the original color of fresh roselle. Color produced by ‘FRESH’ was considered the reference color both in ground and liquid form. In both ground and liquid form, the color of roselle produced by vacuum-drying was closest to the color of fresh roselle (Table 1). This is most probably related to vacuum-drying being based on the rule of producing a vacuum to reduce pressure below vapor pressure of the water and then pressure is decreased around the samples to be dried. During oven-drying and sun-drying, samples were exposed to a continuous flow of hot steam or air [21] which led to discoloration and degradation of the material. Different intensities in red color were observed between drying methods. In liquid form, ‘FRESH’ and ‘VACUUM’ produced light red-yellow color while ‘FREEZE’, ‘OVCOM’, ‘OVLAB’, and ‘SUN’ produced dark red-
blue color. However, in ground form, ‘FREEZE’, ‘OVCOM’, and ‘OVLAB’ produced more lighter red-yellow than ‘FRESH’, ‘VACUUM’ and ‘SUN’ (dark red-yellow). Furthermore, in ground form, freeze-dried roselle calyx had higher redness while sun-dried roselle calyx had higher yellowness. However, in liquid form, ‘FRESH’ was highest both in redness and yellowness. Results related to color of the dried roselle are in accordance with those reported by Juhari et al. [22] and Prachayawarakorn et al. [23], who reported that the main factors affecting color change of materials during drying process were drying time, drying temperature, and loading capacity. The color of roselle that was measured in liquid form showed similar tendencies (Table 1). The processes behind the color changes can be breakdown of anthocyanins, change in anthocyanin color caused by changes in pH, Maillard reactions, and caramelization [24,25].

### Table 1. Physicochemical properties in the fresh and dried roselle (Hibiscus sabdariffa L.) calyx and filtrate.

| Roselle calyx | FRESH | VACUUM | FREEZE | OVCOM | OVLAB | SUN | Significance |
|---------------|-------|--------|--------|-------|-------|-----|--------------|
| Moisture content (%) | 89.4 ± 0.1<sup>a</sup> | 12.9 ± 0.1<sup>c</sup> | 9.6 ± 0.2<sup>d</sup> | 9.5 ± 1.4<sup>d</sup> | 8.9 ± 0<sup>d</sup> | 15.6 ± 0.1<sup>b</sup> | *** |
| Water activity (A<sub>ω</sub>) | 1.000 ± 0<sup>a</sup> | 0.605 ± 0<sup>c</sup> | 0.414 ± 0 | 0.482 ± 0<sup>d</sup> | 0.395 ± 0<sup>e</sup> | 0.727 ± 0<sup>b</sup> | *** |
| Color value - measured in ground form | | | | | | | |
| L* | 34.7 ± 0.4<sup>b</sup> | 35.2 ± 1.1<sup>b</sup> | 38.9 ± 0.3<sup>a</sup> | 39.2 ± 0.4<sup>a</sup> | 39.1 ± 0.4<sup>a</sup> | 38.8 ± 0.5<sup>a</sup> | *** |
| a* | 4.5 ± 0.3<sup>d</sup> | 4.5 ± 1.0<sup>d</sup> | 11.8 ± 0.4<sup>a</sup> | 10.5 ± 0.2<sup>b</sup> | 7.2 ± 0.6<sup>c</sup> | 4.8 ± 0.4<sup>d</sup> | *** |
| b* | 5.9 ± 0<sup>bc</sup> | 6.4 ± 0.5<sup>b</sup> | 7.7 ± 0.1<sup>a</sup> | 7.8 ± 0.2<sup>a</sup> | 5.8 ± 0.1<sup>c</sup> | 8.2 ± 0.5<sup>a</sup> | *** |
| Filtrate | | | | | | | |
| Total soluble solids (%) | 0.4 ± 0.1<sup>f</sup> | 1.1 ± 0.1<sup>e</sup> | 3.8 ± 0<sup>a</sup> | 3.5 ± 0<sup>b</sup> | 3.1 ± 0.2<sup>c</sup> | 2.8 ± 0<sup>d</sup> | *** |
| Color value - measured in liquid form | | | | | | | |
| L* | 43.1 ± 0.1<sup>b</sup> | 45.0 ± 0.1<sup>a</sup> | 35.6 ± 0.1<sup>d</sup> | 35.5 ± 0.1<sup>d</sup> | 35.6 ± 0.2<sup>d</sup> | 36.2 ± 0<sup>c</sup> | *** |
| a* | 20.9 ± 0.2<sup>a</sup> | 16.2 ± 0.2<sup>b</sup> | 6.3 ± 0.1<sup>e</sup> | 7.1 ± 0<sup>d</sup> | 6.1 ± 0.1<sup>e</sup> | 8.8 ± 0.1<sup>c</sup> | *** |
| b* | 17.5 ± 0.2<sup>a</sup> | 15.6 ± 0.1<sup>b</sup> | 5.9 ± 0<sup>d</sup> | 6.1 ± 0.1<sup>d</sup> | 6.0 ± 0<sup>d</sup> | 7.1 ± 0<sup>c</sup> | *** |

Values in a row not marked with the same letter are significantly different, Student’s t-test (p < 0.05). *** Indicates significance at p < 0.001. FRESH = Fresh roselle calyx; VACUUM = Vacuum-dried roselle calyx; FREEZE = Freeze-dried roselle calyx; OVCOM = commercial scale oven-dried roselle calyx; OVLAB = lab scale oven-dried roselle calyx; SUN = sun-dried roselle calyx.

#### 2.2. Volatile Compounds

A total of 74 volatile compounds, consisting of terpenes (18), esters (16), aldehydes (12), ketones (11), furans (6), alcohols (6), phenols (3), and an acid (1), and a lactone (1) were identified in the samples (Table 2). The major volatile compounds (by peak size: methyl acetate, ethyl acetate, 2-butanol, 2-methylbutanol, 3-methylbutanol, pentanal, hexanal, heptanal, methyl hexanoate, limonene, 1-pentanol, p-cymene, α-terpinolene, octanal, 2,6,6-trimethylcyclohexanone, 6-methyl-5-hepten-2-one, nonanal, isoterpinolene, furfural, benzaldehyde, linalool, phenylethanal, 1-phenylethanone, α-terpineol, azulene, δ-cadinene, p-cresol, α-calamorene, and phenol) were found in all samples, but in varying levels.

A Principal Component Analysis (PCA) was carried out using the peak areas relative to dry matter content exhibiting significant (p < 0.05) variation (Figure 1). The first principal component (PC1) explained 46% of the variance while PC2 explained 27% of the variance. Overall, ‘FRESH’ is rather different from dried samples, regardless of the drying method, being characterized by high levels of many terpenes and some aldehydes and esters (mainly branched chain). Terpenes are typical plant volatiles. Twelve of the 18 detected terpenes were significantly higher in ‘FRESH’ and they have odor descriptors like fruity, floral, pine/woody, and spice (Table 2). The fresh roselle had also high levels of some fruity esters (methyl 3-methylbutanoate, methyl pentanoate, and 3-methylbutyl butanoate) and most of the detected aldehydes which among other are described as having green and citrus odors. This corresponds well with Ramirez-Rodrigues [26] who studied aroma profiles of hot and cold infusion of fresh and dried roselle.
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Figure 1. PCA (A) scores and (B) loadings plots of based on average significant $(p < 0.05)$ peak areas of volatile compounds for fresh and dried roselle (Hibiscus sabdariffa L.) calyx samples.
Table 2. Volatile compounds (peak areas relative to dry matter content) identified in the fresh and dried roselle (Hibiscus sabdariffa L.) calyx using different drying methods.

| Compounds        | Calculated RI a | Reference RI | ID b | Odor Description      | FRESH | VACUUM | FREEZE | OVCOM | OVLAB | SUN | Significance |
|------------------|-----------------|--------------|------|------------------------|-------|--------|--------|-------|-------|-----|-------------|
| **Terpenes**     |                 |              |      |                        |       |        |        |       |       |     |             |
| β-Myrcene        | 1154            | 1170         | Standard | Musty, fruity, lemon, spice, woody d | 10534 | 989   | 542    | 0     | 683   | 0   | ***         |
| Limonene         | 1179            | 1200         | Standard | Citrus, fruity, green d  | 39828 | 4057  | 9728   | 9807  | 11478 | 5197 | ***         |
| 1,8-Cineole      | 1187            | 1193         | Standard | Camphor, minty, pine, liquorice, mentholic d | 16625 | 869   | 421    | 1003  | 458   | 0   | ***         |
| p-Cymene         | 1256            | 1261         | Standard | Lemon, fruity, sweet, herbal d | 15767 | 2188  | 36723  | 34849 | 30681 | 4158 | *           |
| α-Terpinolene    | 1270            | 1297         | Standard | Woody, fruity, sweet, pine d | 11089 | 1227  | 4305   | 4218  | 4825  | 1415 | ***         |
| Isoterpinolene   | 1385            | 1331         | Literature | Woody, pine, citrus e | 4447  | 1104  | 920    | 6762  | 3073  | 4642 | ***         |
| trans-Linalool oxide | 1426          | 1438        | Literature | Floral, creamy, earthy, green c | 321   | 5711  | 3316   | 8425  | 0     | 5514 | *           |
| Nerol oxide      | 1460            | 1485         | Standard | Oily, flowery d | 1280  | 0     | 814    | 3038  | 1777  | 0   | *           |
| Camphor          | 1509            | 1498         | Literature | Camphor, green, leafy | 3211  | 1477  | 833    | 7072  | 3582  | 3802 | n.s.        |
| Linalool         | 1528            | 1534         | Standard | Citrus, herbal, flowery c | 548431| 6071  | 5819   | 9672  | 17309 | 22827| ***        |
| α-isophorone     | 1583            | 1591         | Literature | Woody, champor, musty e | 9145  | 376   | 2461   | 1185  | 0     | 1403 | ***        |
| 1-p-menthen-9-al | 1602            | 1620         | Literature | Spice, herbal d | 647   | 3897  | 3007   | 7325  | 4224  | 7277 | n.s.        |
| Aromadendrene    | 1627            | 1628         | Standard | Sweet, dry d | 2063  | 0     | 976    | 0     | 2126  | 0   | n.s.        |
| α-Bisabolene     | 1653            | 1702         | Literature | Berry, spicy, citrus d | 7398  | 0     | 433    | 0     | 370   | 0   | ***        |
| α-Terpineol      | 1683            | 1682         | Literature | Pine, lillac, woody, floral e | 31865 | 2742  | 5374   | 7928  | 6826  | 10288| ***        |
| Azulene          | 1724            | 1746         | Literature | Medicinal d | 35528 | 4193  | 2433   | 3614  | 28692 | 7563 | ***        |
| α-Cadinene       | 1735            | 1749         | Literature | Thyme, medicinal, woody c | 15947 | 1516  | 1944   | 1798  | 2683  | 2979 | ***        |
| α-Calacorene     | 1896            | 1904         | Literature | Woody c | 1104  | 90    | 452    | 261   | 520   | 358 | ***        |
| **Esters**       |                 |              |      |                        |       |        |        |       |       |     |             |
| Ethyl acetate    | 864             | 867          | Standard | Pineapple c, fruity d | 164108| 168821| 5750   | 63801 | 188847| 48039| n.s.       |
| Methyl acetate   | 807             | 810          | Literature | Fruity, solvent-like d | 14252 | 442728| 13023  | 243853| 132356| 290587| **        |
| 2-Methylpropyl acetate | 999        | 1017         | Standard | Fruity, flowery, strong, banana, pear d | 1631  | 17130 | 0     | 1005  | 1905  | 0   | ***        |
| 3-Methylbutyl acetate | 1111       | 1112         | Literature | Sweet, banana, fruity, green e | 602   | 29269| 715    | 1998  | 5342  | 21192| **        |
| Pentyl acetate   | 1165            | 1172         | Literature | Herbal d | 1834  | 1415  | 0     | 1772  | 658   | 8173 | n.s.       |
| Hexyl acetate    | 1266            | 1293         | Standard | Fruity, Herbal c | 1041  | 1140  | 0     | 0     | 0     | 13768| *          |
| Phenethyl acetate| 1797            | 1795         | Standard | Rose, floral, fruity, sweet d | 0     | 2889  | 0     | 0     | 0     | 17139| **        |
Table 2. Cont.

| Compounds               | Calculated RI a | Reference RI | ID b | Odor Description             | FRESH | VACUUM | FREEZE | OVCOM | OVLAB | SUN | Significance |
|-------------------------|-----------------|--------------|------|------------------------------|-------|--------|--------|-------|-------|-----|--------------|
| Other esters            |                 |              |      |                              |       |        |        |       |       |     |              |
| Methyl 2-methylpropanoate | 907             | 910          | Literature | Fruity, floral e             | 3557  | 10362  | 906    | 736   | 783   | 0   | ***          |
| Methyl 2-methylbutanoate | 991             | 1000         | Literature | Apple, fruity c               | 799   | 6133   | 0      | 0     | 0     | 0   | ***          |
| Methyl 3-methylbutanoate | 1003            | 1011         | Literature | Fruity, apple d              | 13640 | 4149   | 12     | 0     | 120   | 0   | ***          |
| Methyl pentanoate       | 1076            | 1086         | Standard | Sweet, ethereal, apple d     | 1606  | 0      | 0      | 397   | 101   | 0   | *            |
| Methyl hexanoate        | 1176            | 1196         | Standard | Fruity, fresh, sweet c       | 1515  | 1691   | 213    | 3057  | 662   | 1762 | *            |
| 3-Methylbutyl butanoate | 1236            | 1256         | Literature | Fruity, apple, spicy, buttery e | 40922 | 0      | 0      | 1709  | 0     | 0   | ***          |
| Methyl octanoate        | 1375            | 1401         | Standard | Orange e                      | 0     | 299    | 174    | 586   | 466   | 4871 | **           |
| Methyl nonanoate        | 1471            | 1481         | Literature | Coconut c                     | 0     | 0      | 87     | 847   | 377   | 1622 | ***          |
| Methyl salicylate       | 1759            | 1797         | Standard | Peppermint c                  | 602   | 447    | 176    | 1020  | 356   | 480  | n.s.         |
| Aldehydes               |                 |              |      |                              |       |        |        |       |       |     |              |
| 2-Methylpropanal        | 787             | 789          | Literature | Green, pungent, burnt, malty d | 0     | 11900  | 5142   | 7050  | 12196 | 6909 | *            |
| 2-Methylbutanal         | 893             | 896          | Standard | Fruity, almoned, toasted, malty, green d | 14846 | 37241  | 5803   | 12035 | 29298 | 22709 | n.s.         |
| Pentanal                | 948             | 968          | Standard | Almond, malty, pungent d      | 46310 | 8401   | 7752   | 22431 | 29236 | 20175 | n.s.         |
| Hexanal                 | 1068            | 1087         | Standard | Grassy c                      | 100121| 5753   | 16053  | 59056 | 27115 | 46659 | ***          |
| Heptanal                | 1172            | 1192         | Standard | Fatty, citrus, rancid c       | 15731 | 946    | 1235   | 6669  | 3076  | 5293  | ***          |
| 2-Hexanal               | 1196            | 1205         | Literature | Apple, green, leaf c          | 14741 | 0      | 3260   | 1753  | 2056  | 1767  | ***          |
| Octanal                 | 1279            | 1311         | Standard | Orange peel, pungent f        | 20491 | 772    | 1263   | 4275  | 2202  | 6123  | **           |
| Nonanal                 | 1379            | 1402         | Standard | Fatty, citrus, green c        | 75446 | 3183   | 5419   | 23189 | 12695 | 20012 | ***          |
| Decanal                 | 1480            | 1511         | Standard | Green, waxy, floral, tallow d | 5368  | 3360   | 388    | 1733  | 1233  | 2793  | *            |
| Benzaldehyde            | 1503            | 1537         | Standard | Almond c                      | 53396 | 22030  | 8172   | 11396 | 30133 | 49355 | *            |
| Phenylethanal           | 1622            | 1636         | Literature | Honey, sweet c                | 2733  | 3822   | 1344   | 2384  | 2867  | 4030  | n.s.         |
| Ketones                 |                 |              |      |                              |       |        |        |       |       |     |              |
| 2-Butanone              | 877             | 881          | Standard | Ether-like d, fruity e        | 100594| 13176  | 2816   | 5570  | 9444  | 8845  | ***          |
| 2-Pentanone             | 1103            | 1023         | Literature | Fruity, wine, woody e         | 318   | 9732   | 0      | 3618  | 5085  | 0     | n.s.         |
| 4-Methyl-3-penten-2-one | 1112            | 1113         | Literature | Minty d                       | 378   | 1054   | 388    | 2804  | 1936  | 1599  | **           |
| 2-Heptanone             | 1168            | 1189         | Standard | Soap f, blue cheese f         | 6000  | 613    | 69     | 2061  | 712   | 6591  | ***          |
| 6-Methyl-2-heptanone    | 1223            | 1228         | Literature | Camphoroous e                 | 3796  | 547    | 190    | 1666  | 1397  | 5859  | **           |
| 2-Octanone              | 1276            | 1283         | Literature | Earthy, woody, herbal, yeasty e | 2081 | 230    | 0      | 1204  | 0     | 15423 | *            |
| 2,6,6-Trimethylcyclohexanone | 1304            | 1333         | Literature | Pungent d, honey, citrus f    | 3322  | 441    | 414    | 3570  | 1345  | 3164  | ***          |
| 6-Methyl-5-hepten-2-one | 1329            | 1339         | Standard | Mushroom, earthy, woody, rubber d | 17970 | 3421   | 4008   | 12375 | 7464  | 26764 | ***          |
| 3-Octen-2-one           | 1381            | 1392         | Literature | Earthy, spicy, herbal e       | 327   | 0      | 0      | 0     | 0     | 2768  | ***          |
| 2-Undecanone            | 1579            | 1580         | Literature | Waxy, fruity, pineapple e     | 551   | 0      | 0      | 440   | 256   | 2597  | ***          |
| 1-Phenylethanol         | 1632            | 1645         | Literature | Almond, floral d, musty c     | 6241  | 2515   | 1165   | 2683  | 2196  | 3052  | ***          |
Table 2. Cont.

| Compounds                              | Calculated RI a | Reference RI | ID b | Odor Description | FRESH | VACUUM | FREEZE | OVCOM | OVLAB | SUN | Significance |
|----------------------------------------|-----------------|--------------|------|------------------|-------|--------|--------|-------|-------|-----|--------------|
| Furans                                 |                 |              |      |                  |       |        |        |       |       |     |              |
| 2-Methylfuran                          | 865             | 877          | Literature | Ether-like, chocolate d | 478   | 929    | 0      | 932   | 821   | 1317 | n.s.         |
| 2-Pentylfuran                          | 1218            | 1229         | Literature | Green bean c, pungent f | 6330  | 2120   | 1877   | 31274 | 6815  | 53767*** |
| 5-Isopropenyl-2-methyl-2-vinyltetrahydrofuran | 1227            | 1253         | MS    | Fresh, forest, grassy c | 407   | 3770   | 3579   | 25511 | 9142  | 4485 | ***          |
| Furfural                               | 1443            | 1458         | Standard | Bread, almond c      | 5093  | 379572 | 185077 | 565607 | 442585 | 380159 *** |
| 2-Acetylfuran                          | 1486            | 1497         | Standard | Balsamic c           | 606   | 22074  | 6933   | 35757 | 29182 | 32601*** |
| 5-Methyl-2-furfural                    | 1554            | 1560         | Standard | Almond, caramel, burnt sugar c | 1710  | 9757   | 2555   | 53826 | 23679 | 24234*** |
| Alcohols                               |                 |              |      |                  |       |        |        |       |       |     |              |
| 2-Methyl-1-propanol                    | 1089            | 1100         | Standard | Ethereal, whiny e   | 765   | 12669  | 497    | 619   | 4286  | 9570 | ***          |
| 3-Methylbutanol                       | 1199            | 1222         | Standard | Fusel, pungent, ethereal, banana e | 50370 | 29394  | 576    | 6440  | 20607 | 33726*** |
| 1-Pentanol                             | 1248            | 1274         | Standard | Alcohol, pungent, fruity, balsamic c | 6211  | 1651   | 935    | 2987  | 2475  | 10508** |
| 1-Hexanol                              | 1348            | 1372         | Standard | Resin, flowery, green c | 118601 | 3188   | 1663   | 0     | 3238  | 72258*** |
| 1-Nonanol                              | 1639            | 1640         | Literature | Fatty, green e     | 915   | 0      | 0      | 0     | 0     | 3752 | ***          |
| Phenethyl alcohol                      | 1894            | 1932         | Standard | Honey, spice, rose, lilac c | 0    | 3761   | 0      | 0     | 585   | 5563 | ***          |
| Phenols                                |                 |              |      |                  |       |        |        |       |       |     |              |
| p-Cresol                               | 1884            | 1902         | Literature | Medicinal, phenol, smoke c | 2201  | 1163   | 4553   | 6744  | 6671  | 8057 | n.s.         |
| Phenol                                 | 1976            | 1987         | Literature | Phenolic, medicinal d | 6872  | 1163   | 609    | 826   | 499   | 762 | ***          |
| p-Ethylguaiacol                        | 2005            | 2008         | Literature | Spice, clove e      | 0    | 329    | 1707   | 646   | 1459  | 2854 | *            |
| Pentanoic acid                         | 1647            | 1685         | Literature | Sweaty, pungent, sour, cheesy, beefy d | 0    | 744    | 0      | 0     | 0     | 0 | *            |
| γ-Butyrolactone                        | 1610            | 1617         | Literature | Caramel, sweet c    | 461   | 2709   | 305    | 1072  | 1037  | 2902 | **          |

a The retention indices (RIs) of volatiles were calculated as the retention time of the volatiles normalized to the retention times of adjacently eluting n-alkanes (C6-C22); b Identification method: ‘Standard’: retention time and mass spectrum confirmed by authentic standard run on the same system, ‘Literature’: linear retention index is close to retention indices from Flavornet/Pherobase/NIST Webbook/PubChem for DB-wax capillary GC column, ‘MS’: mass spectrum agrees with mass spectrum in database; c Odor description based on Flavornet; d Odor description based on Pherobase; e Odor description based on The Good Scents Company; f Odor description based on Odor.org.uk; *,**,*** indicate significance at p < 0.05, p < 0.01, and p < 0.001, respectively; n.s. means no significant difference between the samples.
All of the dried samples showed lower levels of the above-mentioned compounds, but significantly higher levels of five of the six furan compounds found. These compounds have odor descriptors like bread, almond, burnt sugar, and green/herbal and are often found in heat treated or roasted foods [27,28]. Chen et al. [29] found that thermal processing through air-drying produced caramel-like aroma, and this was also reported when drying roselle calyces [9,29,30]. Furans like furfural and 5-methyl-2-furfural can be formed through sugar degradation during heat treatment [28]. This formation can be accelerated by low moisture content (water activity 0.3–0.7 [31]). In our study, the dried samples had water activities in this range. Two terpenes (trans-linalool oxide and nerol oxide) were highest in most of the dried samples, probably due to oxidation during drying [29].

Among the dried samples, the sun-dried differ by having significantly higher levels of certain ketones (2-heptanone, 2-octanone, 2-undecanone, 6-methyl-2-heptanone, 6-methyl-5-hepten-2-one, and 3-octen-2-one), esters (hexyl acetate, phenethyl acetate, methyl hexanoate, methyl octanoate and methyl nonanoate), and alcohols (pentanol and phenethyl alcohol). These compounds are not normally related to drying or heat treatment but rather indicate some degree of fermentation. This is most probably due to the earlier mentioned haze phenomenon, which made the sun drying slow and incomplete, and the findings for sun-dried samples in this study should therefore not be generalized.

2.3. Microstructure

Microstructural evaluation can elucidate product changes during the drying process and record how well-preserved the tissue is after the treatments. Figure 2 shows scanning electron micrographs of the surface of fresh and dried roselle calyx. The epidermis of the fresh calyx consists of thick-walled, tightly packed, and well-organized cells (Figure 2a), whereas the drying processes introduced various physical changes in the material (Figure 2b–f). The structural integrity of plant foods is mainly attributed to the primary cell wall, the middle lamella, and the turgor pressure generated within cells by osmosis. During most food processing operations, the turgor pressure is lost imparted by the disruption of the plasma membrane and vacuolar membrane, leading to cellular shrinkage.

However, no particular shrinkage was observed in the epidermis of the freeze-dried samples (Figure 2b). The surface was mostly smooth, the cell structure was well preserved and overall the sample appeared to have a more fresh-like quality. This is due to the fact that the samples were frozen before being dried, and the water was subsequently sublimated. The final structure of freeze-dried roselle is thus formed during freezing. This results in a porous structure, which will influence texture and dehydration capacity.

The microstructure of roselle was clearly affected by the vacuum-drying treatment (Figure 2c) as seen by the dehydrated, shrunken appearance of the epidermis cells. On the other hand, the cell layer appeared intact and individual cells could be identified as a result of this treatment. An explanation for the fairly well-preserved microstructure could be the formation of polymeric material resulting in increased thickness of the middle lamella between cells [32]. It was also observed that the texture of vacuum-dried roselle was more elastic and stretchable compared to the other dried samples.

Oven-drying extensively affected the roselle tissue structure and led to cell wall disruption, deformation, and folding (Figure 2d,e), probably introduced by the high drying temperature and velocity. This result is in agreement with other studies on apples [33,34]. The highest degree of tissue disruption and cell collapse was found in sun-dried roselle samples (Figure 2f) probably due to the long exposure.

The differences in microstructure may well explain some of the differences in color since a rough surface would be expected to have a lighter hue (lightness), while the original shape and structure (i.e., in fresh and freeze-dried) would intensify and deepen the color. This point is in accordance with Yousif et al. [35] who studied physical properties of vacuum-microwave-air-dried sweet basil. Further, the loss of turgor pressure during the drying processes will introduce color changes to the tissue, since the color of the roselle
epidermis is due to water-soluble pigments located in the vacuole as well as lipid-soluble pigments located in the cytoplasm.

![Image of scanning electron micrographs](image_url)

**Figure 2.** Scanning electron micrographs of the surface of roselle (*Hibiscus sabdariffa* L.) calyx samples after different treatments at a magnification of 500×. Bar = 10 µm: (a) fresh; (b) freeze-dried; (c) vacuum-dried; (d) commercial scale oven-dried; (e) lab scale oven-dried; (f) sun-dried.

### 3. Materials and Methods

#### 3.1. Sample Preparation

Fresh roselle (*Hibiscus sabdariffa* L.) calyx of the UMKL cultivar (obtained from HERBagus Sdn. Bhd, Kepala Batas, Malaysia) was chosen for the study. Samples used were harvested at a fully mature stage. The confirmation of plant species identification was based on taxonomic descriptions and photographic illustrations by botanist Dr. Shamsul Khamis from the Institute of Bioscience (UPM Serdang, Malaysia). The fresh samples were manually sorted and washed thoroughly under running water to remove dirt and other extraneous matter. The excess water was drained and then the samples were weighed and kept in a chiller at 4 °C (less than 48 h) for further use.
3.2. Drying Experiments

Four different drying methods were compared in this study. Appropriate drying times and temperatures were determined in a preliminary experiment. The dried samples were kept in aluminum coated zip-lock packaging and stored at room temperature until further analysis. All drying experiments were performed in triplicate.

Oven-drying was carried out by (1) using a commercial scale oven (‘OVCOM’) in HERBagus Sdn. Bhd. Penang, Malaysia, and (2) using a laboratory scale oven (‘OVLAB’) in Faculty of Food Science and Technology, Universiti Putra Malaysia, Selangor, Malaysia. For commercial scale, a programmable oven model Box Oven 36 Tray (Kimah Industries Suppliers (M), Perai, Malaysia) was used. The drying oven was equipped with an air ventilation outlet, temperature controller, and timer switch which allow the user to select the required drying temperature and adjust the time of drying. During the experiment, a perforated drying tray (90 cm × 58 cm) and (56 cm × 56 cm) (Kimah Industries Suppliers (M), Perai, Malaysia) was used. The air was circulated from the above and the bottom of the drying tray. For laboratory scale, fresh roselle calyx samples were dried on a perforated drying tray (73 cm × 65 cm) using a programmable oven 400W (Smoke Master Model SMA-112, Hanagi Seisakusho Co., Ltd., Toda, Japan). This oven was also equipped with a temperature control function and the air was circulated from the above and the bottom of the drying tray. The samples for both commercial and laboratory-scale were dried as a monolayer at 50 °C for 12 h at a constant air flow of 2 m s⁻¹. The relative humidity of the ambient air (30 °C) was around 60–68%. During the drying process, the temperature of the drying air was recorded by a portable thermometer (EL-EnviroPad-TC, Corintech, Hampshire, United Kingdom) via an attached thermocouple probe (Figure 3). The oven was switched on for 30 min before the drying process to stabilize the temperature.

For freeze-drying (‘FREEZE’), samples were prepared as in 3.1 and then frozen in a freezer at −24 °C. Drying was carried out over 48 h in a freeze dryer (Freeze dry System LABCONCO Freezone 18®, Kansas City, MO, USA) at −40 °C and a pressure of 3.3 × 10⁻³ mbar.

Vacuum-drying (‘VACUUM’) was conducted using a vacuum-drying oven with a vacuum pump (Model VD 53, WTB Binder, Berlin, Germany) at 50 °C for 26 h at a pressure of 28 mbar. Then, 400 g of sample were spread as a monolayer on the aluminum expansion
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racks and were dried in the vacuum chamber. The temperature was recorded as described at oven-drying. The oven was switched on for 30 min before the drying process to stabilize the temperature.

Sun-drying ('SUN') was carried out in a monolayer on a tray directly under the hot sun (<45 °C) for 19 h. The temperature was recorded as above. The relative humidity during sun drying was around 59–70%.

3.3. Physicochemical Analysis

Fresh and dried roselle calyx samples were ground for 2 min with high speed using a blender (Panasonic MX-M1011, Petaling Jaya, Malaysia) and mixed thoroughly. The ground samples were kept in aluminum coated zip-lock packages and stored at room temperature until further analysis. Moisture content was determined in triplicate using an air-oven method at 105 °C until constant weight [36]. The water activity was measured using a water activity meter (Aqualab Series 3 TE, Decagon Device, Inc, Pullman, WA, USA). Samples were measured in triplicate at 25 ± 1 °C. The samples were placed in a glass petri dish (7.4 cm diameter) and read by a Minolta Chroma Meter CR-300 Series 2° (Konica Minolta Sensing Americas, Inc., NJ, USA) observer through the bottom of the petri dish. The colorimeter was calibrated using a standard Minolta calibration plate. Triplicate measurements were recorded using the \( L^* \), \( a^* \), \( b^* \) system.

An extract was prepared from 4 g of ground roselle mixed with 40 mL of tap water and soaked for 30 min. The extract was filtered using filter paper (S&S folded filters, Schleicher & Schuell, Chicago, IL, USA, 320 mm). Total soluble solids were determined in triplicate using an LCD Digital Bench Model Refractometer (HI96801, Hanna Instrument Inc., Nusfalau, Romania) and color was measured as described above.

3.4. Volatile Compounds

All analyses were carried out in triplicate as reported by Juhari et al. [30]. Whole roselle (fresh or dry) was ground for 2 min using a blender (KRUPS Speedy PRO, Group SEB Nordic AS, Ballerup, Denmark), and 10 g of ground roselle was mixed with 40 mL of tap water. The compound 4-methyl-1-pentanol (1 mL of a 5 mg L\(^{-1}\) solution) was added as internal standard. Each sample was placed in a gas washing flask (300 mL, 7.5 cm diameter) together with a magnetic stirrer to agitate the sample during volatile extraction. Volatile compounds were collected on a Tenax-TA trap connected to the flask’s outlet. The trap contained 200 mg of Tenax-TA mesh size 60/80, density of 0.37 g mL\(^{-1}\), Buchem bv, Apeldoorn, The Netherlands. The samples were equilibrated to 40 °C ± 1 °C in a circulating water bath and then purged with nitrogen (100 mL min\(^{-1}\)) for 30 min. Water was removed from the traps using a flow of dry nitrogen (100 mL min\(^{-1}\) for 10 min). The Tenax-TA traps were then capped and stored at 5 °C before analysis by gas chromatography-mass spectrometry.

The trapped volatiles were desorbed using an automatic thermal desorption unit (TurboMatrix 350, Perkin Elmer, Shelton, CT, USA). Primary desorption was carried out by heating the trap to 250 °C with a flow (50 mL min\(^{-1}\)) of carrier gas (H\(_2\)) for 15.0 min. The stripped volatiles were trapped in a Tenax TA cold trap (30 mg held at 5 °C), which was subsequently heated at 300 °C for 4 min (secondary desorption, outlet split 1:10). This allowed for rapid transfer of volatiles to a gas chromatograph-mass spectrometer (GC-MS, 7890A GC-system interfaced with a 5975C VL MSD with Triple-Axis detector from Agilent Technologies, Palo Alto, CA, USA) through a heated (225 °C) transfer line. Separation of volatiles was carried out on a DB-Wax capillary column (30 m long \times 0.25 mm internal diameter, 0.50 µm film thickness). The column pressure was held constant at 2.4 psi resulting in an initial flow rate of 1.4 mL min\(^{-1}\) using hydrogen as carrier gas. The column temperature program was: 10 min at 30 °C, from 30 °C to 240 °C at 8 °C min\(^{-1}\), and finally 5 min at 240 °C. The mass spectrometer was operating in the electron ionization mode at 70 eV. Mass-to-charge ratios between 15 and 300 were scanned. Volatile compounds were identified by probability-based matching of their mass spectra with those of a commercial
database (Wiley275.L, HP Product no. G1035A) using the software program, MSDChem-
station (Version E.02.00, Agilent Technologies, Palo Alto, CA, USA). Volatile compound
identification was confirmed by comparison with retention indices (RI) of authentic refer-
ence compounds or retention indices in the literature. Since the study included fresh as
well as dried roselle calyces, the results from volatile analyses are presented as peak areas
relative to dry matter content.

3.5. Microstructural Analysis

Tissue pieces were arranged on aluminum stubs on double-sided tape with the epi-
dermis upwards, and coated with a layer of gold using a Sputter Coater SCD 005 (Bal-Tec
Company, Buffalo Grove, IL, USA) for the dried roselle calyx, whereas no coating of a
sample was done on fresh roselle calyx. The dried samples were examined using secondary
electron (SE) detector while the fresh samples were examined using backscattered electron
(QBSD) detector in a scanning electron microscope, SEM (Leo 1455VP Variable Pressure,
Cambridge, UK) operating at an accelerating voltage of 20 kV. Micrographs were taken at a
magnification of 100×, 500× and 1000×. The microstructural analysis was carried out in
triple.

3.6. Data Analysis

All data from the physicochemical analyses and volatile compound analysis were
analyzed by one-way analysis of variance (ANOVA) using the software JMP 13 (SAS
Institute Inc, Cary, NC, USA). Post hoc calculations using Student’s t-test were used for
multiple comparisons. Multivariate analysis was applied to GC-MS data to evaluate the
variation between the fresh and dried roselle samples. PCA is a multivariate projection
method designed to extract and visually display the systematic variation in the data matrix
of the volatile compounds, making it possible to include many statistical variables at
the same time. PCA was performed using the Latentix software (LatentiX 2.0, Latent5,
Copenhagen, Denmark). Analyses were carried out on the average of significant (p < 0.05)
peak areas and the data were auto-scaled.

4. Conclusions

All the tested drying methods, except sun-drying, reduced moisture content and water
activity to a safe level (water activity = 0.605 or lower). When extracts were prepared,
it turned out that the extract based on freeze-dried roselle had the highest value of TSS.
Furthermore, the color after vacuum-drying was the one closest to the color of fresh samples,
both when evaluated in ground and liquid form. All drying methods reduced terpenes,
some aldehydes, and esters which are expected to contribute to typical fruity, floral, spicy,
and green odors. At the same time furan compounds increased. These compounds are most
likely causing the caramel-like aroma that has been reported in dried roselle. Among the
dried samples, the sun-dried stood out as having higher levels of some ketones, alcohols,
and esters. This was possibly due to unintended fermentation processes. Drying was found
to significantly affect microstructure, but freeze-drying preserved the cell structure best.
The different drying methods applied had different effects on quality, and the present study
may therefore serve as a tool for the choice of the most appropriate drying techniques to be
used for the production and further commercialization of dried roselle calyx.

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