Development of Mist Dispersion Technology for Aomori Hiba Oil

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A technology that disperses Hiba oil with mist-like consistency is being developed for new applications of Hiba oil such as for preventing allergic reactions in the living environment and pest control in agriculture in order to heighten the added value of the Hiba oil, a special product of Aomori Prefecture.

In this research conducted as part of that development, methods of quantitatively evaluating the dispersibility and dispersion durability of Hiba oil mist generated from an experimental mist generator were examined. Acceptable results were obtained for Hiba oil and Hinokitiol in evaluation using multipoint humidity measurement and evaluation using the color reaction caused by ferric chloride.

Key words: Aomori Hiba Oil, hinokitiol, mist dispersion, multipoint humidity measurement, colorimetry

1. INTRODUCTION

Aomori Prefecture accounts for about 80% of Japan's total stock of Hiba wood as supplied from its prefectural tree. Hiba oil is obtained by steam-distilling the scrap wood resulting from the lumbering of hiba. Aomori Industrial Research Center has clarified the excellent effects of Hiba oil such when used in insect control, antibacterial, and antifungal applications[1,2]. However, Hiba oil is not widely used in antimicrobial substances and mothballs, but has only been commercialized in such daily goods as bath oil and shampoo because it is not readily or uniformly dispersed in water.

Conversely, chemically synthesized medicines and agricultural chemicals have been used to control mold, bacteria, and viruses that damage agricultural products and cause infantile asthma and atopic dermatitis. However, the safety of food has become a growing concern among consumers due to the problems posted by unregistered agricultural chemicals and outbreak of Bovine Spongiform Encephalopathy (BSE) and avian influenza in recent years. Therefore, there is now an urgent need to improve the living environment and agriculture through the efficient use of natural materials[3].

This research was conducted to develop technology of efficiently dispersing Hiba oil, which is a natural material, in the form of a mist for such new applications as preventing allergic reaction in the living environment and pest control in agriculture in order to heighten the added value of Hiba oil, a special product of Aomori Prefecture. In this paper, results of examining the methods of quantitatively evaluating the dispersibility of Hiba oil mist is reported.

2. EXPERIMENTAL METHOD

2.1 Aomori Hiba oil

Aomori Hiba oil is yellowish to brownish oily liquid obtained by steam-distilling the sawdust and chips generated by cutting Aomori Hiba (*Thujopsis dolabrata* Sieb. *et* Zucc. var. *hondai* Makino) wood into lumber. Aomori Hiba oil contains 1% hinokitiol as the principal antibacterial component (Table 1, Fig. 1)[1].

| Constituent | | Content (% | |
|-------------|------------------|------------|--|
| Neutral oil | Thujopsene | 50~60 | |
| | Cedrol | 5~10 | |
| Acidic oil | Hinokitiol | 1~2 | |
| | β-Dolabrin | 1~2 | |
| | Citronellic acid | 1~2 | |
| etc. | | 24~42 | |

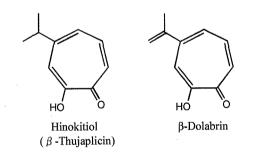


Fig. 1 Structure of Hinokitiol and β -Dolabrin

2.2 Production of Hiba oil preparation and mist generator

The preparation of Hiba oil was made by mixing Aomori Hiba oil, mono-lauric acid decaglyceryl (Nikko Chemicals, DECAGLYN 1-L), and pure water in a weight ratio of 1:1:98, and stirring the mixture for five minutes at 4500rpm with an internal shear force type of mixer (M Technique, Clearmix CLM-0.8S) (Table 2). Moreover, an ultrasonic-type mist generator that generates mist particles about 2µm in diameter by using 3-MHz ultrasonic elements was made for trial purposes and its mist dispersion performance was checked.

Table 2 Hiba oil preparation

| Composition | wt% | |
|---------------------------|-----|--|
| Hiba oil | 1 | |
| Decaglycerol mono-laurate | 1 | |
| Water | 98 | |

2.3 Quantitative evaluation with multipoint humidity measurement

Multipoint humidity measurement was examined as a method of quantitatively evaluating the mist dispersion. A measurement system that simultaneously measures both temperature and humidity with temperature-humidity IC sensors (Sensirion, STH15) was produced experimentally. These electrostatic capacity type of high-polymer sensors enable measurement with accuracy of ±2.0% RH. The sensors were located at four points (at heights of 0cm, 25cm, 50 cm, and 75cm) one meter away from the mist generator in an enclosed space (170cm × 60cm × 120cm, Fig.2).

Measurement was initiated immediately upon closing the measuring space, followed by the mist generation five minutes later for 15-minute atomization. The measuring space was opened 15 minutes after stopping the atomization, and the measurement continued under the same conditions was ended five minutes after opening the space (Fig.3). Humidity distribution was also compared between pure water and the Hiba oil preparation by generating mist of them with the generator.

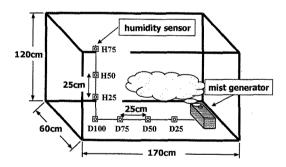


Fig.2 Measurement environment and points

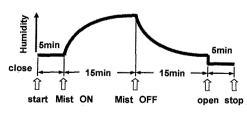


Fig.3 Humidity measurement profile

2.4 Quantitative evaluation based on ferric chloride color reaction

Hinokitiol, which is the principle antibacterial component of Hiba oil, forms a complex with metal ions. Therefore, a method of quantitatively evaluating mist dispersion state based on color reaction caused by the Hinokitiol complex formation was examined. Two or more sheets of cut filter paper with ferric chloride applied are located with the mist generator in the enclosed space in the same way as that for the multipoint humidity measurement, and the degree of coloring on the filter paper specimens was measured with a colorimeter (Minolta, CR-200). Color difference ΔE between the uncolored part and colored part on each specimen was calculated using the L*a*b* color A water-soluble Hinokitiol preparation system[5]. (Table 3) was adjusted until the Hinokitiol density became 5000ppm to produce complexes sufficient to obtain a clear evaluation result, and then atomized for three hours.

Test filter paper was cut into $30\text{mm} \times 30\text{mm}$ specimens and ferric chloride solution (Wako Pure Chemical Industries, Ltd.) at a 40% concentration was applied on the specimens and allowed to dry. Then, half of the thus treated surface of each specimen was covered with tape as a control. Seven or six specimens thus treated were attached to both sides of thin plates positioned vertically and horizontally, respectively.

Table 3 Water-soluble hinokitiol preparation

| Composition | wt% |
|---------------------------|-----|
| Hinokitiol | 3 |
| Decaglycerol mono-laurate | 20 |
| Ethanol | 30 |
| Glycerin | 5 |
| Water | 42 |

2.5 Examination of mist generation method

The three main types of the humidifier (ultrasonic, heating, and vaporizing) were compared to determine the most suitable type for the mist dispersion of Hinokitiol. A device experimentally developed for Hiba oil mist dispersion, the Toshiba KA-C3S, and Toshiba KA-D55X were used as the ultrasonic type, heating type, and vaporizing type, respectively. The quantitative evaluation based on the color reaction caused by ferric chloride was used for the comparison purposes. Filter paper specimens with ferric chloride applied were located 50 cm away from the mist generator. The water-soluble Hinokitiol preparation adjusted to achieve a Hinokitiol density of 1000ppm was atomized.

3. RESULTS AND DISCUSSIONS 3.1 Quantitative evaluation with multipoint humidity measurement

The results of humidity measurement for atomization of pure water and Hiba oil preparation were shown in Figures 4 and 5. There was little difference in the humidity depending on the measurement position (height) in both results, thus suggesting that the mist was dispersed uniformly in the measuring space.

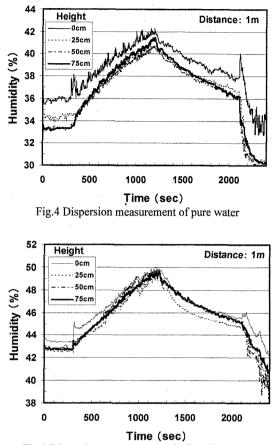


Fig.5 Dispersion measurement of Hiba oil preparation

3.2 Quantitative evaluation based on ferric chloride color reaction

Photographs of filter paper specimens tested when arranged vertically are shown in Figure 6 and a graph drawn by plotting the color differences converted from the degree of coloring is shown in Figure 7.

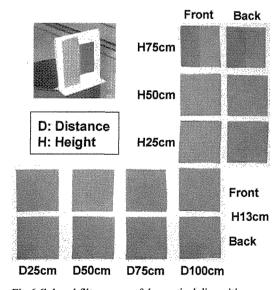


Fig.6 Colored filter paper of the vertical disposition

Theses results show a tendency toward deeper coloring of the filter paper specimens on the side opposite the mist generator than those on the same side as the generator. The results also show a remarkably deep coloration of specimens at a height of 75 cm. This colorization was attributed to irregular diffusion due to the direction of the fan of in the mist generator.

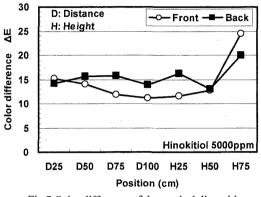


Fig.7 Color difference of the vertical disposition

Photographs of filter paper specimens tested when arranged horizontally are shown in Figure 8 and a graph drawn by plotting the color differences converted from the degree of coloring is shown in Figure 9. When compared, the specimens were found to be colored to almost the same degree regardless of whether they were attached to the upper side or the underside, thus suggesting uniform diffusion. Although the degree of coloration was only remarkably high for specimens 25 cm away from the generator, this finding was attributed to the direct exposure of these specimens to the mist generated by the generator. This test suggests a uniform diffusion of Hinokitiol in the measuring space.

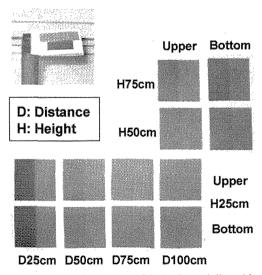


Fig.8 Colored filter paper of the horizontal disposition

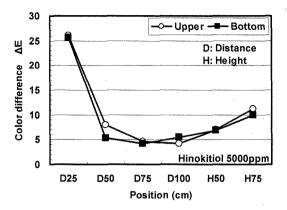


Fig.9 Color difference of the horizontal disposition

3.3 Examination of mist generation method

Sheets of filter paper tested with the ultrasonic-type, heating-type, and vaporizing-type humidifiers are shown in Figure 10, and the data measured with the L*a*b* color system and the calculated color differences (ΔE) from the uncolored surfaces are listed in Table 4. The vaporizing-type and heating-type were expected to Hinokitiol more uniformly diffuse than the ultrasonic-type because of their high humidification capability. However, the color difference measurement results of the test using ferric chloride color reaction showed that the ultrasonic type diffuses Hinokitiol most uniformly. From this finding, it became clear that a ultrasonic-type was most suitable for diffusion of the Hinokitiol, an antibacterial component.



Vaporizing

Ultrasonic

Fig.10 Colored filter paper of each mist generation method

Table 4 Evaluation of coloring by the L*a*b* color system

| | L* | a* | b* | ΔE |
|-------------------|-------|-------|--------|------|
| None mist (basis) | 72.60 | 10.20 | 74.86 | - |
| Vaporizing | 72.69 | 9.23 | 73.19 | 1.93 |
| Heating | 70.44 | 8.00 | .71.16 | 4.82 |
| Ultrasonic | 68.76 | 9.23 | 67.32 | 8.52 |

4. CONCLUSION

As a result of multipoint humidity measurement by using temperature-humidity sensors to quantitatively evaluate the dispersibility and dispersion durability of Hiba oil mist in an encloseed space, the same diffusion state of Hiba oil as that of pure water was confirmed. Sheets of cut filter paper with ferric chloride applied were also arranged in the measuring space and color reaction caused by metal complex formation was examined to evaluate the dispersibility of Hinokitiol, which is the principle antibacterial component of Hiba Coloration was observed on the specimens on the oil. front and back of the measurement location, thus showing that Hinokitiol was sufficiently diffused in the measuring space. In addition, the ultrasonic, heating, vaporizing-type humidifiers were compared for use in mist generation and it was clarified that the ultrasonic-type was most suitable for the diffusion of Hinokitiol.

Hiba oil can be dispersed with mist-like consistency by using a Hiba oil preparation and ultrasonic-type mist generator such as experimentally produced in this research, and new applications of Hiba oil such as for the control of insects and diseases can be expected in various fields of the living environment and agriculture.

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