Research of essential oil extraction efficiency improvement from *Thujopsis dolabrata* var. *hondae* using the underwater shock wave

Ayumi Takemoto * and Shigeru Itoh **

*Shock Wave and Condensed Matter Research Center, Kumamoto University,
2-39-1 Kurokami, Kumamoto City, Kumamoto 860-8555, Japan

Tel: +081-96-342-3288-305 Fax: +081-96-342-3293 e-mail: tkmt@mech.kumamoto-u.ac.jp

**Okinawa National College of Technology,

905, Henoko, Nago City, Okinawa, 905-2192, Japan,

Tel: +081-980-55-4000, Fax: +081-980-55-4012, e-mail: itoh@okinawa-ct.ac.jp

The underwater shock wave selectively destroys the tracheid of the conifer by the effect of the spalling destruction, and forms the extraction road of an internal tree sap. The shock wave that is the instantaneous high pressure reaches the entire cell, and the effects are expected more than usual static pressures. As for the essential oil obtained by steaming distilling Hiba (*Thujopsis dolabrata* var. *hondae*) sawn wood, a variety of physiology revitalizations like the antibacterial activity and the effect etc. of insecticide of Hinokitiol that is the principal ingredient are admitted. As a preprocessing, treating the underwater shock wave to Hiba sawn wood, the essential oil is expected to be able to be extracted more highly effective than a usual steam distillation. In this research, the increasing of the amount of Hinokitiol included in the essential oil by the shock wave loading is reported.

Key words: Shock Wave, Instantaneous High Pressure, Spalling Destruction, Essential Oil, Hiba

1. INTRODUCTION

The essential oil of Hiba (*Thujopsis dolabrata* var. *hondae*) of which the principal ingredient is Hinokitiol is known most as an essential oil obtained from the plant that grows naturally in Japan. As for Hinokitiol, wide physiology revitalization of an insecticidal action has been reported. For instance, anti-bacterium revitalization to plant pathogens such as *Valsa ceratosperma*, bacillus such as methicillin-resistant *Staphylococcus aureus*^[1], insecticidal action on Yamato termite (*Reticulitermes speratus*) and acaricidal action on Kenagacodani (*Trypohagus putrescentiae*)^[2], and so on. The yield amount of high performance material Hiba essential oil is almost 1.0-1.5mL from the Hiba wood 100g.^[1] The improvement of the extraction efficiency leads to effective use for a high performance material. Authors experimented on the improvement of the extraction using the underwater shock wave loading to *Cryptomeria japonica* D. Don (the Japan cedar, Sugi) that was the conifer as well as Hiba in a past research.^[3] It is an effect of the spalling destruction that the shock wave brings. Then, authors aim at the improvement of the Hinokitiol extraction using the underwater shock wave loading for Hiba sawn wood in this research.

2. SPALLING DESTRUCTION

The shock wave transmits in the material at the speed that exceeds speed of sound. The shock wave divides into the transparent wave and the reflected wave on the change side of the density difference as shown in Fig. 1. The expansion wave is a speed below speed of sound. The reflected wave caused on a high density side brings the pull stress from negative pressure. And, the pull stress causes exfoliation effect, that is, spalling on the surface of the density difference. This high-speed

destruction phenomenon is called spalling destruction. It seems that the spalling destruction is caused by the density difference between the cell wall, the bubble in the anatomy, and the cytoplasm when the shock wave passes the plant cell.^[4]

Fig.1 Schematic view of the spalling destruction mechanism

In the conifer, the pit membrane that exists on the tracheid is destroyed by the shock wave, and the pass road is formed. Fig.2 shows the scanning electron microscope (SEM) photograph by the low vacuum scanning electron microscope (Carry Scope JCM-5700, JEOL Ltd.) of Hiba wood. The pit membrane lines up on the tracheid. The conifer is composed of the passage of water that is called the tracheid, by the connection of short tubes, and carries the water sucked up from the root to the leaf. The tube connects with the other tube by the pit membrane to pass the water. The pit membrane causes the effect of the spalling destruction according to the shock wave load, and is destroyed selectively. As a result, the pass road is formed in the conifer. And it is expected that the essential oil is expected to be extracted efficiently by the pass road.^[3]

Fig.2 Scanning electron microscope photograph, Structure of Hiba

3. EXPERIMENT

3.1 Experimental set-up

Fig. 3 shows the outline of the experimental set-up. The sawn wood obtained from Hiba that had been deforested in Aomori Prefecture was used for the sample. The detonating fuse (The Japan Carlit Ltd. Soc 6,308m/s) was set from the sample in the distance of 100mm. Experimental set-up was sunk in the experiment water tank, and detonated with the electric detonator (percussion cap made of Asahi Chemical Industrial Co., Ltd. No.6). Strength of the shock wave pressure is requested by the distance between the detonating fuse and the sample.^[5] In this experiment, sample was loaded 4 times by the underwater shock wave of 120MPa.

Fig.3 Schematic view of the experimental set-up
“d” shows distance between the detonating fuse and the sample.

The scanning electron microscope of Hiba sawn wood was observed before and behind the shock wave loading. The scanning electron microscope used the carry scope JCM-5700 (JEOL Ltd.). The picture was taken in the low vacuum mode.

In addition, differences by the presence of shock wave loading of the Hinokitiol content of the extracted essential oil were measured by using the spectrophotometer and compared. The UV-3600 UV-VIS-NIR Spectrophotometer (Shimadzu Corporation) was used for the absorbance measurement.

3.2 Steam distillation

The essential oil has been extracted from Hiba sawn wood by using the small steam distilling apparatus after the shock wave loading. Hiba sawn wood for the extraction used 517g, and distilled water for the extraction used 3,000ml. 20 minutes were required from the heating to the extraction beginning, and more 40 minutes were required to the extraction end. The extracted oil is 0.85ml. In the same way, the extracted oil from untreated Hiba sawn wood of 527g is 0.80ml.

4. RESULTS

4.1 SEM observation

The SEM photograph of untreated after extraction sample after the the shock wave loading is shown in Fig.4 and the SEM photograph of shock wave loading treated sample are shown in Fig.5 and Fig.6. Each sample was taken of a SEM photograph after steam distillation. As for the untreated Hiba sawn wood, there is no crack on the tracheid (Fig. 4). As for the shock wave treated Hiba sawn wood, a lot of cracks are caused on the tracheid as shown in Fig.5. In addition, the pit membrane has flaked off from the tracheid as shown in Fig.6. These are thought the effect of the spalling destruction by the shock wave, and will function as a pass road at steam distillation.

Fig.4 Untreated after extraction sample without crack on the cell wall

Fig.5 Detonating shock wave treated after extraction with cracks on the cell wall

Fig.6 Stripping bordered pit on detonating shock wave treated sample after extraction

4.2 Contents of Hinokitiol

The Hiba essential oil contains active ingredient Hinokitiol with a variety of physiology revitalizations^[6]. The absorbance was measured by using the spectrophotometer outside purple visible to examine the Hinokitiol content of the Hiba essential oil extracted. Three kinds of Hiba essential oils were used for measurement. One is the extracted oil from the shock wave treated Hiba sawn wood, next is from the untreated Hiba sawn wood, and the last one is the essential oil on the market (TREE OF LIFE Co.,Ltd. Lot number 1, Hinokitiol content 2.08%). 99.5% Dehydrated ethanol was used for the solvent. Essential oils were diluted to 4×10^{-4} times with the solvent respectively.

Fig. 7 shows the absorbance curve in about 320 nm, where the Hinokitiol crystal (made of the Aomori Prefectural Industrial Technology Research Center) shows a peak. The absorbance of untreated Hiba oil is almost equal to Hiba oil on the market, and the absorbance of the shock wave treated Hiba oil is obviously stronger than those of others.

Fig.7 Comparing of extraction rate shock wave treated samples and untreated extraction

- Extracted oil from the shock wave treated Hiba sawn wood
- Essential oil on the market
- - - - - Extracted oil from the untreated Hiba sawn wood

5. CONCLUSION

As a result of the experiment, signs of the spalling destruction were confirmed on the tracheid of Hiba sawn wood by the shock wave load. In addition, the

absorbance curve indicates that the Hinokitiol content of the extracted oil from shock wave treated Hiba sawn wood have a relatively high Hinokitiol content than the extracted oil from untreated Hiba sawn wood and the essential Hiba oil on the market. On the other hand, the qualitative evaluation is necessary because the Hinokitiol analogue that shows the absorbance near Hinokitiol is included in the Hiba essential oil in the future. That is, the shock wave processing is useful for the improvement of the essential oil extraction efficiency.

Repeating the shock wave treatment is necessary for the extraction from the tree cell. It is unsuitable for processing it repeated because the shock wave load with a detonating fuse is a batch processing. Continuous processing is indispensable to obtain a lot of essential oils by industrial. Therefore, it will be necessary to replace the detonating fuse and to conduct the extraction experiment by the repetition processing of the shock wave treatment with a continuous processor in the future.

6. ACKNOWLEDGMENT

This work was supported by Grants-in-Aid for Scientific Research (21923006) by Japan Society for the Promotion of Science(JSPS). And the authors thank Dr. T. Okabe, Aomori Prefectural Industrial Technology Research Center for his providing samples.

7. REFERENCES

- [1] Yatagai, M. "Aromas of forests and trees, their characteristics and functions" Journal of Japan Association on Odor Environment 38(6), 428, 2007
- [2] Okabe, T., Morita, Y., Inamori, Y., Narita, K. and Yamamoto, R. "About the application of Aomori Hiba oil and the application and the Aomori Hiba material recycling circulation system of the Aomori Hiba oil" Wood Preservation, 32(5), 225-231, 2006
- [3] Takemoto, A. and Itoh, S., "Non-heating extractive from *Cryptomeria japonica* D. Don (Sugi) using the underwater shock wave" Transactions of the Materials Research Society of Japan, 32(4), 1071-1073, 2007
- [4] Takemoto, A., Kuroda, K., Iyama, H. and Itoh, S. "On the destruction of the cell wall of plants and its mechanism by the shock wave" The International Journal of Multiphysics, 2(2), 165-170, 2008
- [5] Murata, K., Takahashi, K., Kato, Y., Nagano, S. and Itoh, S. "Measurement of the underwater shock wave produced by the underwater explosion of detonating cord (in Japanese edition)" Abstracts of Japan Explosives Society in spring, 73-74, 1997.
- [6] Morita, Y., Sakagami, Y., Okabe, T., Ohe, T., Inamori, Y., Ishida, N. "The mechanism of the bactericidal activity of hinokitiol" Biocontrol Science, 12(3), 101-110, 2007

(Received May 15, 2010; Accepted January 10, 2011)