The Effect of Underwater Shock Waves on Steam Distillation of *Alpinia zerumbet* leaves

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*Alpinia zerumbet* (Pers.) B.L. Burtt & R.M. Smith is an aromatic perennial plant, distributed widely in the Ryukyu Islands, Japan. It has been extensively studied because its essential oil has a variety of biological functionalities. Underwater shock waves consist of instantaneous high pressures that penetrate entire plant cells and selectively destroy the cell walls of leaves and stems by spalling destruction. Therefore, it is expected that exposing leaves to shock waves as a pre-processing step draws more effective extraction of essential oil by subsequent steam distillation. In this study, we prepared *A. zerumbet* materials that were subjected to underwater shock wave pretreatment, and evaluated its effect on the leaves for steam distillation. With shock wave loading, multiple cracks in the cell walls were observed by scanning electron microscopy. Moreover, the extracted volume of volatile compounds increased with the number of shock wave processing. In particular, the concentrations of camphor and \( p \)-cymene in the vapor phase increased more than four times compared with these of untreated leaves. Underwater shock wave processing selectively and effectively destroyed the fiber and/or cell structures. The headspace gas chromatography/mass spectrometry indicated that the volatile components within the structures were easily volatilized.

Key words: *Alpinia zerumbet*, Essential Oil, Shock Wave, Underwater Shock Wave

1. INTRODUCTION

*Alpinia zerumbet* (Pers.) Burtt & Smith is an aromatic perennial plant that is widely distributed throughout the tropical and sub-tropical regions of the world, and grows freely from southern Kyushu to the Ryukyu Islands in Japan. *A. zerumbet* is known as “sannin” in the local Okinawa dialect and “getto” in Japanese. It is used to flavor and wrap “mochi” rice cakes and its leaves are sold as herbal tea. The volatile components of essential oils, obtained by hydrodistillation from *A. zerumbet* leaves, were investigated in 1965 [1]. Teruya, Ikema and co-workers worked on the antifungal, antimicrobial, insecticidal and antioxidant activities of essential oils in the 1980s–90s [2–5]. In recent research, new functions of the essential oil were discovered, including chemoprotective effects against H\( _2 \)O\( _2 \)-induced DNA damage in cultured human leukocytes [6] and advanced glycation end product inhibitors [7]. However, the essential oil is very expensive because large quantities of the leaves are required to obtain relatively small amounts of the oil (0.04 - 0.07%) [1,2].

Underwater shock waves consist of instantaneous high pressure that penetrates the entire plant cell and selectively destroys the cell walls of leaves and stems by spalling destruction. In shock wave-treated Hiba sawn wood, multiple cracks were generated on the tracheid and the pit membrane flaked off the tracheid through spalling destruction. It was concluded that these cracks function as a permeation pathway during steam distillation [8]. Therefore, the application of underwater shock wave treatment to the leaves and stems of *A. zerumbet* as a pre-processing step is expected to enable more effective extraction of essential oil by subsequent steam distillation. In this study, the extracts from *A. zerumbet* materials that were untreated or subjected to underwater shock wave pretreatment were analyzed, and the effect of underwater shock waves on the leaves of *A. zerumbet* for steam distillation was evaluated.

2. EXPERIMENTAL

2.1 Experimental Setup

The processing equipment for pretreatment of leaves and stems of *A. zerumbet* by an underwater shock wave was developed. A diagram of the equipment is shown in Fig. 1. The processing equipment consists of a vessel, high-voltage power supply unit and a high-voltage regulator.

The high-voltage supply device generates a voltage of 2 to 4 kV in an 800 \( \mu \)F capacitor and the underwater shock wave is generated momentarily in the vessel by electrical discharge. This processing equipment can continuously produce an instantaneous high pressure, which constitutes an impulse force that crushes the leaves and the stems. *A. zerumbet* leaves and stems were placed into a
silicone tube (i.d. 40 mm, o.d. 50 mm) and subjected to the instantaneous high pressure load produced by the underwater shock wave.

2.2 Underwater Shock-Wave Loading

The leaves and the stems of *A. zerumbet* were supplied by Green Plan Shinjo Ltd. in Okinawa. The leaves and stems were finely cut and dried at 45°C to maintain a moisture content of 10 to 20%. The samples were subjected to the high pressure produced by the underwater shock wave four times. It is presumed that the pressure produced by the underwater shock wave generating equipment was about 40 MPa. The effect on the leaves was evaluated before and after the shock wave loading by analyzing the cell structure in scanning electron microscope (SEM) images and the volatile components extracted from the leaves.

SEM images of *A. zerumbet* leaves were observed before and after shock wave loading, using a S-3000N instrument (Hitachi High-Technologies Corp., Tokyo, Japan). The SEM images were taken in the high vacuum mode.

2.3 Analysis of Volatile Components of Leaves

The leaves and the stems of *A. zerumbet* were observed using the headspace gas chromatography-mass spectrometry (HS-GC/MS) method. One gram of leaves were sealed with a septum in a 20 ml gas-tight vial. The sample vial was pressurized above the capillary column head pressure with carrier gas using a headspace (HS) sampler (TurboMatrix HS-40, PerkinElmer, Inc., MA, USA) and heated to 60 °C during 22 min to equilibrate with vapor-phase extraction. The pressurized vapor phase including volatile compounds was then transferred to the GC/MS (QP-2010 Plus, Shimadzu Co., Kyoto, Japan) with an injection time of 0.05 min. A ZB-WAX Plus column of 60 m length, 0.32 mm i.d., and 0.5 µm thickness (Phenomenex Inc., Torrance, CA, USA) was used. The carrier gas was helium. The GC oven temperature program was as follows: 40 °C held for 3 min, increased at 5 °C/min to 165 °C, 10 °C/min to 220 °C and held for 3 min. The injector and detector temperatures were set at 250 °C. The mass range was scanned from 30 to 600 amu. The control of the GC/MS system and the data peak processing were carried out using Shimadzu's GC/MS solution software, version 2.7. The volatile components were identified by comparing their retention times and mass fragmentation patterns with those of standards and an MS library. Quantitative determinations of essential oil components were carried out based on peak area measurements. Additionally, differences in the volatile component content of the leaves caused by the shock wave loading were determined using peak area measurements.

3. RESULTS AND DISCUSSION

3.1 SEM Observation

SEM images of the cell structures in *A. zerumbet* leaves before and after shock wave loading at 3.0 kV, 3.6 kJ, are shown in Figs. 2 and 3, respectively. A regular structure was observed before shock wave loading, whereas after shock wave loading, the cell structures were crushed and multiple cracks were generated on the cell walls by the instantaneous high pressure. These are believed to be the effect of spalling destruction by the shock wave, and it is expected that these cracks function as a permeation pathway during steam distillation. Thus, the underwater shock wave reached the entire cell and selectively destroyed the cell walls in the leaves and stems.

![Fig. 1 Diagram of the processing equipment for underwater shock wave pretreatment of *A. zerumbet* leaves and stems.](image1)

![Fig. 2 SEM image of the cell structure of *A. zerumbet* leaves before shock wave loading.](image2)

![Fig. 3 SEM image of the cell structure of *A. zerumbet* leaves after shock wave loading at 3.0 kV, 3.6 kJ. Each arrow shows cracks that were generated on the cell walls by the instantaneous high pressure.](image3)
3.2 Major Volatile Components of Leaves in the Vapor Phase Extraction

The major volatile components of dried leaves and stems of *A. zerumbet* treated by underwater shock wave under various conditions are shown in Fig. 4. Quantitative determinations of volatile components were carried out based on peak area measurements of GC/MS spectra for untreated leaves. Shock wave processing was performed once, twice, or three times on dried leaves and each measurement result was compared with that of untreated leaves. The major volatile components detected in the vapor-phase extraction were α-pinene, camphene, limonene, 1,8-cineol, α-phellandrene, β-cymene, hexanal, β-pinene, myrcene and camphor. These components represented 94.3% of the detected total components. The most of volatile compounds sharply increased with the number of the applied shock wave loading times. In particular, the concentrations of camphor and β-cymene were increased more than four times compared with these of untreated leaves. 1,8-Cineol was not observed in the untreated dried leaves, so the concentration ratio of 1,8-cineol was calculated based on the peak area of spectrum for leaves processed with a single shock wave loading.

These results show that fiber and cell structures were split and crushed by shock wave processing and the volatile components inside the structure were easily volatilized by heating because the underwater shock wave penetrated the entire tissue and selectively destroyed the cell walls of leaves and stems.

Conversely, the concentration of hexanal was not affected by shock wave loading. It is thought that hexanal was distributed to various regions in the leaves and volatilizes easily irrespective of the existence of cracks on the cell walls.

Although α-phellandrene and myrcene oozed out and adsorbed on the surface of the leaves like other compounds when the leaves were finely cut and dried at low-temperature, the concentration didn’t rise after the first shock wave loading. Since it was thought that these compounds of more quantity eliminated by the first shock wave loading and it was hard to volatilize because these compounds distributed deep inside the cells, these concentration once decreased. However, these compounds were volatilized after having subjected to the high pressure repeatedly and increased again.

Thus, the application of an underwater shock wave treatment as a preprocessing step to the leaves and stems of *A. zerumbet* is expected to enable more effective extraction of essential oil by subsequent steam distillation.

4. CONCLUSION

The results of SEM observation confirm that multiple cracks were formed by shock wave loading in the cell walls of *A. zerumbet* leaves, caused by spalling destruction. The most of vapor-phase concentrations of extracted components increased with the number of applied times shock wave processing. Moreover, the concentrations of some components increased more than four times compared with untreated leaves. These results show that shock wave loading is useful for improving essential oil extraction efficiency. Repeated processing is possible using the instantaneous high pressure caused by a momentary electric discharge but it is necessary to continuously handle large quantities of leaves to obtain industrial volumes of essential oils. Therefore, processing equipment for continuously feed of leaves into the underwater shock wave treatment vessel and extraction experiments by repetitive shock wave treatment processing are needed in the future.

5. ACKNOWLEDGEMENT

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6. REFERENCES