

Feasibility of food processing treatment using plasma-based ion implantation method

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Abstract: Plasma-based ion implantation (PBII) is a surface modification method, which applies a negative high-voltage pulse to a sample in plasma. Ions are implanted uniformly by reducing the width of the ion sheath formed near the sample and by aligning the sheath shape more with the sample for complexly shaped samples. This method is considered a potentially effective surface treatment method for various shapes of objects. This study investigated the PBII method to explore its potential use as a food processing device. The research samples were pork belly and cabbage. The investigation results showed decreased bacteria viability, although the sample temperature at the treatment time was less than 40 °C. Additionally, the plasma treatment could not decrease the vitamin C content. Furthermore, fine holes could be created on the sample surface. These results indicate that PBII treatment is usable in food processing to improve the permeability of seasoning solutions while ensuring food safety and nutrient stability.

KEYWORDS: PLASMA-BASED ION IMPLANTATION (PBII), FOOD PROCESS, STERILIZATION, VITAMIN C

1. Introduction

Modern food production requires processing that is safe, free of chemical contamination and without loss of taste or functionality. From a safety standpoint, altering food using microorganisms is a major problem. Generally, heat sterilization is used to prevent food alteration by microorganisms. However, new sterilization technologies, including ultra-high pressure, electrolysis, and plasma treatment, which differ from conventional heat sterilization, are being developed [1-3]. With our concern being plasma treatment, we have focused particularly on the PBII method, which does not use peroxide gas, owing to its harmfulness to the human body [4]. We have pursued its potential as a sterilization device. We have investigated the effects of nitrogen and oxygen gas on heat-resistant spore bacteria [5-7]. It has also been reported that the PBII method effectively sterilizes *Escherichia coli* and *Staphylococcus aureus* [8]. Recently, consumer preferences have become more diverse, and various new food products have been developed, consequently requiring new food processing equipment. Realistically, pressurized thermal sterilization equipment is being used as pressurized thermal cooking equipment. Therefore, this study aimed to explore the potential of PBII treatment equipment as a food processing device.

2. Materials and Methods

In this experiment, food materials were used instead of the microorganisms used in previous studies. Therefore, because of operational problems, the PBII treatment apparatus was used without an external radiofrequency (RF) excitation source.

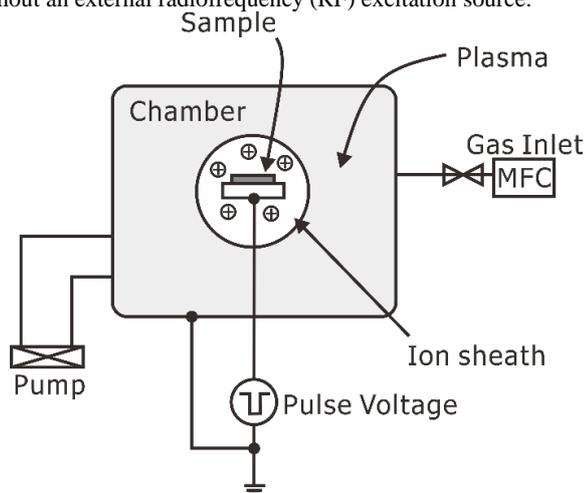


Fig. 1 Schematic of the PBII apparatus.

Fig 1 shows a schematic of the PBII apparatus [9,10]. The treatment chamber is electrically grounded and 200 and 150 mm in height and diameter, respectively. In the experiment, the sample fixed sample plate was placed on a stainless-steel electrode supported by an insulated stainless-steel rod at the center of the vacuum chamber. The chamber's inner temperature during plasma

treatment was measured using the thermolabel for vacuum (VL-40, NiGK Corporation).

The cabbage and pork belly used in the experiment were purchased from Hiroshima City. Samples were cut into approximately 2 cm × 2 cm pieces and dried on a stainless-steel Petri dish in the same procedure as when the microorganisms were sterilized. Table 1 shows a summary of the conditions for PBII treatment.

Table 1: Setting of the PBII apparatus

Items	Value
Supplied gas	O ₂
Gas pressure [Pa]	5
Pulse voltage [kV]	-1 to -10
Exposure time [min]	10

After the plasma treatment, food samples were measured for general viable bacteria counts using standard agar media according to common hygiene practices in Japan. Data were obtained using twelve times analyses, which were expressed as the average ± standard deviation. The sterilization effectiveness was indicated by dividing the number of bacteria after plasma treatment by the number of viable bacteria before plasma treatment to calculate the percentage killed.

Vitamin C concentrations were determined using the 2,4-dinitrophenylhydrazine method. Data were obtained using nine times analyses, which were expressed as the average ± standard deviation.

The observation samples were covered with a thin conductive gold layer using an ion sputtering apparatus (JEOL JFC-1600). Thereafter, the sputtered samples were observed using a scanning electron microscope (JEOL JSM-5610).

3. Results and Discussion

Figs 2 and 3 show the death rate data of viable bacterial counts for cabbage and pork belly after PBII treatment. We previously reported that PBII treatment using RF power could produce sterilizing effects of 6D or higher on *Geobacillus stearothermophilus*, *Escherichia coli*, and *Staphylococcus aureus*.

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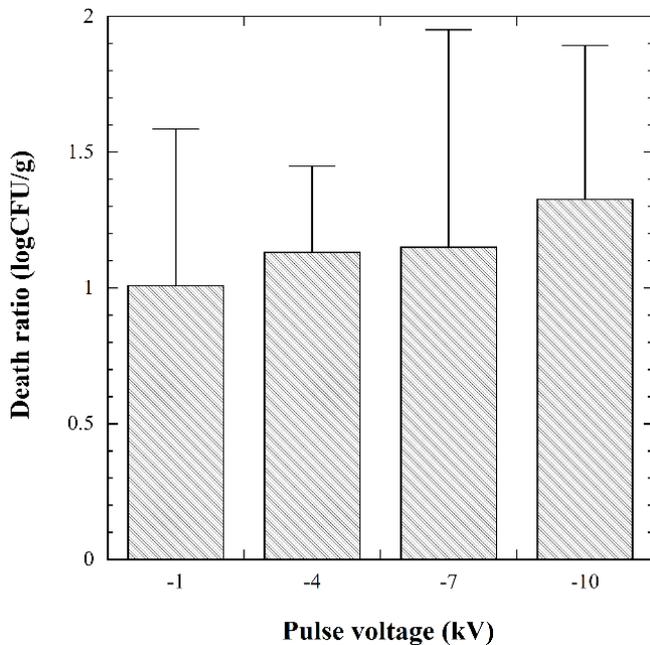


Fig. 2 Death ratio after the PBII-treated cabbage.

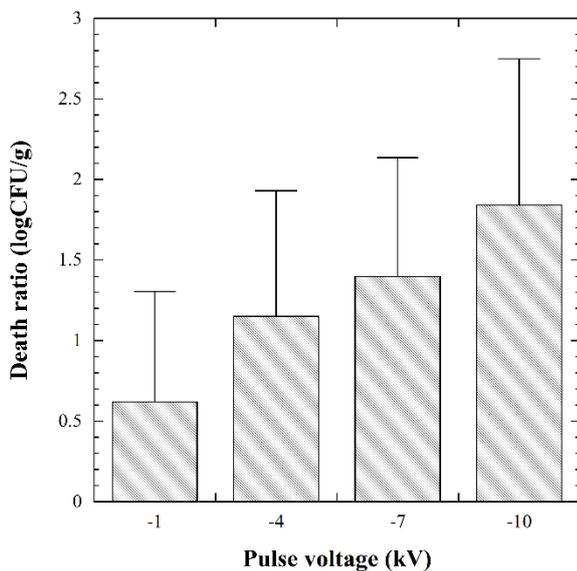


Fig. 3 Death ratio after the PBII-treated pork belly.



Fig. 4 Result of the temperature measurement.

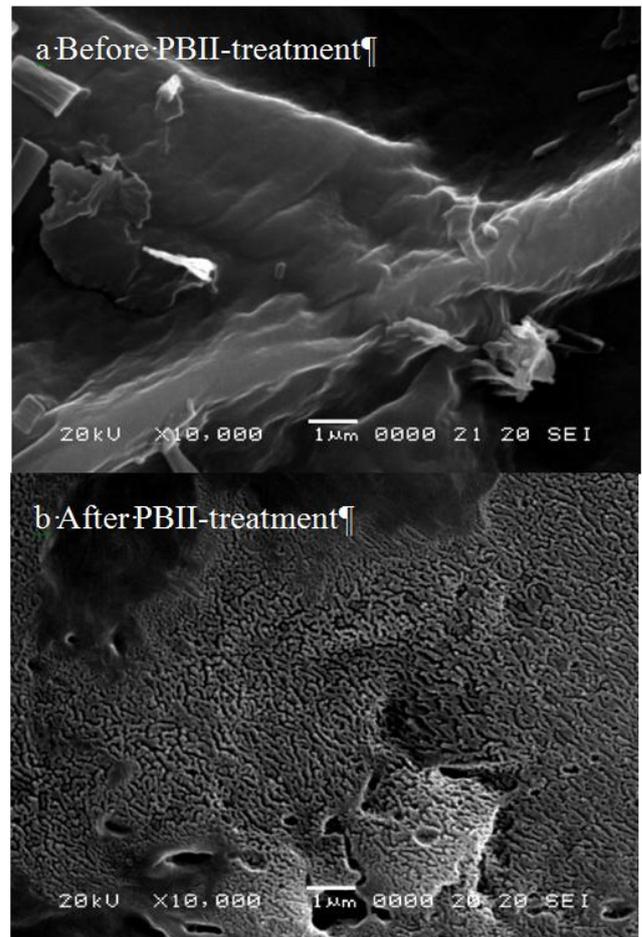


Fig. 5 SEM observation results of cabbage.

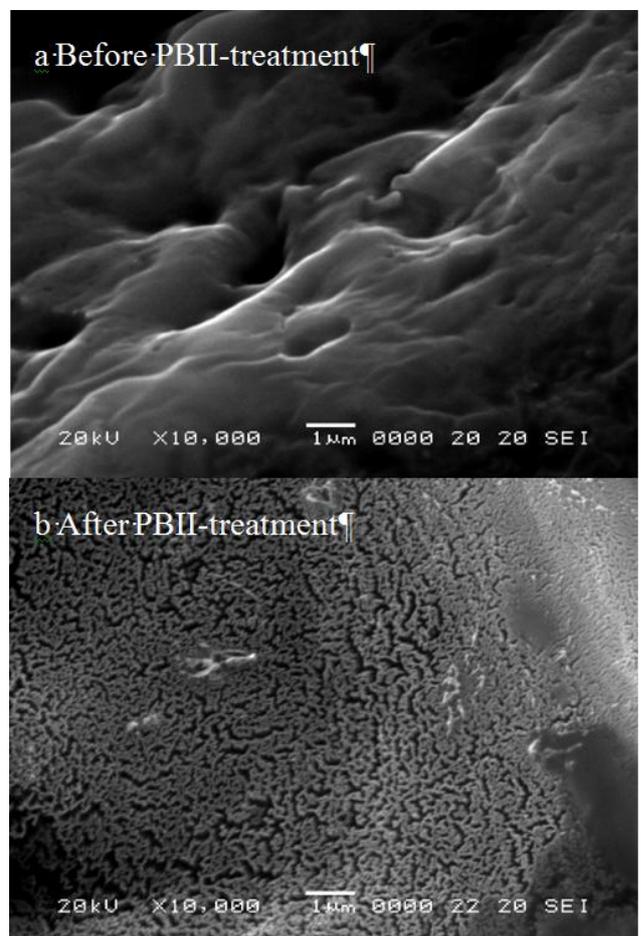


Fig. 6 SEM observation results of pork belly.

However, in this study, the sterilizing effect on cabbage and pork belly was 1.3D and 1.8D, respectively, when both were treated at -10 kV. For this study, the sample is fixed to conductive stainless steel, but the food itself is a material with poor electricity conductivity. Generally, self-ignited arc discharge is unlikely to occur, as well as secondary electron emission from the sample surface. Therefore, when treating food materials, secondary electron emission is less likely compared to the case of metals, and the sterilizing effect will supposedly be higher when RF output is used. Fig 4 shows the chamber's inner temperature measurement results. The thermolabel used in the experiment irreversibly changes the temperature detection area from light yellow to black when the temperature exceeds the indicated value. The results in fig 4 shows that the chamber's temperature was below 40°C even at an applied voltage of -10 kV in the apparatus used in this experiment. For this study, the bacteria sterilization is supposedly caused by physical damage to the bacteria due to collisions between the ions and the bacteria, and not by temperature. Therefore, it can be considered that even at temperatures below 40°C, where sterilization rarely occur, a sterilizing effect of 1.8 D was achieved in the case of pork belly. In contrast, for cabbage, there was almost no increase in the sterilizing effect with an increase in the applied voltage. Supposedly, this is because the cabbage's surface is highly uneven, possibly preventing ions from impinging on it, as well as because the viable bacterial count initially attached to the cabbage is low, around 102 (CFU/g), making it difficult to significantly express the sterilizing effect in numerical terms (See the electron micrograph data for the adhering material on the cabbage surface).

Figs 5 and 6 show the electron micrographs of cabbage and pork belly before and after PBII treatment, respectively. The applied voltage used for PBII treatment was -10 kV, and the electron microscope magnified 10000× in both cases. Although detailed data are not shown, no surface scratches were observed on either cabbage or pork belly at voltages lower than -6 kV, and the number of scratches increased as the applied voltage was increased. This may be due to increased ions impinging on the sample as the applied voltage increased. Also, the difference in conductivity between the cabbage and pork belly may influence the number of ions that collide. As figs 5b and 6b show, the cabbage has a larger area without holes, suggesting that the cabbage was less electrically conductive than the pork belly and that the ion implantation was less efficient in some areas. This could also be the cause of the difference in the sterilizing effect.

Finally, we investigated whether PBII treatment affects the food's nutrient content. In this study, cabbage, known for its high vitamin C content, was used to investigate whether PBII treatment affected vitamin C content. The results are shown in fig 7.

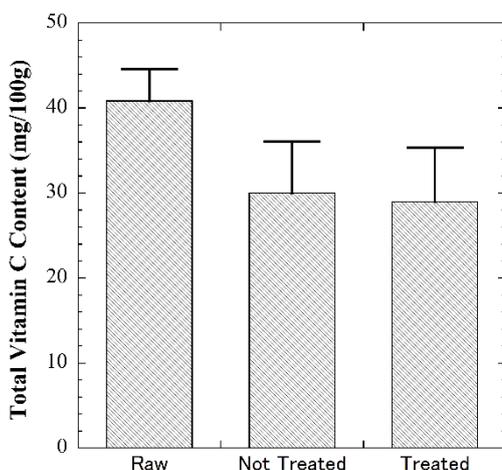


Fig. 7 Effect of PBII treatment on the vitamin C content of cabbage.

In this study, the food sample was dried before PBII treatment. As shown in fig 7, the dried cabbage, indicated by "Not treated," had 25.2% less vitamin C than the raw cabbage, from 40.1 mg

(mg/100g) to 30.0 mg (mg/100g). In contrast, no significant difference was found between the vitamin C content of the dried food samples and the PBII-treated food samplings. Therefore, the PBII treatment itself does not affect the vitamin C content. Generally, immersion in sodium hypochlorite is used when processing raw cabbage. Thus, the total vitamin C content is reported to decrease by 27% [11]. Therefore, future work will probably be in the preparation stage of PBII treatment. This study's results indicate the possibility of piercing the surface of food products with PBII treatment. This result suggests that PBII treatment can improve the permeability of seasoning and enzyme solutions by applying them to the food before cooking. Additionally, the PBII treatment probably has no effect on the food's nutritional components, such as vitamin C. Although data are not shown, no significant change was observed in the texture of the cabbage or pork belly before and after PBII treatment. This is probably due to the shallow depth of the holes generated by the PBII treatment. A softening treatment technology for foods intended for the elderly is necessary. In the future, if the conditions of PBII treatment can be optimized to deepen the wound depth and soften the texture of the food surface, new applications may be possible. Furthermore, if the RF power not used in this study is usable to increase the sample's product temperature, more effective heat sterilization and cooking can be achieved together. These results indicate that the PBII treatment device can potentially be applied as a new food processing device.

4. Conclusion

PBII treatment with oxygen gas was performed on pork belly and cabbage. Consequently, a decreased number of viable bacteria were observed for pork belly, although the temperature of the PBII-treated samples was below 40°C. Additionally, the plasma treatment did not reduce the vitamin C content. Furthermore, microscopic pores could be opened on the sample surface in both cases of pork belly and cabbage. RF power is required for further enhancement of the sterilizing effect. The number of pores on the sample surface increased depending on the applied voltage increment. These results suggest that PBII treatment can simultaneously perform sterilization and food processing if the operating conditions are appropriately determined.

5. Acknowledgment

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6. References

- [1] Jacobs, P. T., S.-M. Lin, in: R.L. Clough, S.W. Shalaby (Eds.), *Irradiation of polymers : fundamentals and technological applications*, American Chemical Society, Washington, DC, 1996, pp. 216-239.
- [2] Kelly-Wintenberg, K., A. Hodge, T. C. Montie, L. Deleanu, D. Sherman, J. Reece Roth, P. Tsai, L. Wadsworth, "Use of a one atmosphere uniform glow discharge plasma to kill a broad spectrum of microorganisms", *Journal of Vacuum Science & Technology A*, 17, 1999, 1539-1544.
- [3] Moreau, S., M. Moisan, M. Tabrizian, J. Barbeau, J. Pelletier, A. Ricard, L. H. Yahia, "Using the flowing afterglow of a plasma to inactivate *Bacillus subtilis* spores: Influence of the operating conditions", *Journal of Applied Physics*, 88, 2000, 1166-1174.
- [4] Yoshida, M., T. Tanaka, S. Watanabe, T. Takagi, M. Shinohara, S. Fujii, "Experimental study on a new sterilization process using plasma source ion implantation with N₂ gas", *Journal of Vacuum Science ; Technology A*, 21, 2003, 1230-1236.
- [5] Hiyama, M., K. Kakugawa, A. Yakushiji, K. Watanabe, R. Matsuda, Y. Tsuchiya, TanakaT., "Optimum conditions for sterilization of *Geobacillus stearothermophilus* spores using plasma-based ion implantation", *ELECTROTECHNICA & ELECTRONICA*, 53, 2018, 199-203.
- [6] Kakugawa, K., S. Umemoto, T. Noda, K. Nosaki, T. Tanaka, K. Vutova, "Effect of the conductivity of the petri dish placed on the

- electrode on the surface layer of the spores in the PBII method", *ELECTROTECHNICA & ELECTRONICA*, 57, 2022, 41-45.
- [7] Fujimura, N., K. Shimono, M. Kubo, H. Noguchi, H. Toyota, K. Kakugawa, T. Tanaka, "Generation of Oxygen Plasma Using Plasma-based Ion Implantation and its Application to Sterilization of Spore-forming Bacteria. (in Japanese)", *IEEJ Transactions on Fundamentals and Materials*, 135, 2015, 373-378.
- [8] Umemoto, S., K. Kakugawa, K. Nosaki, T. Noda, T. Tanaka, K. Vutova, "The sterilization effect of plasma-based ion implantation on prokaryotic microorganisms", *ELECTROTECHNICA & ELECTRONICA*, 57, 2022, 46-50.
- [9] Tanaka, T., T. Hironaka, S. Hayashi, I. Koyama, "Disinfection and sterilization by using plasma-based ion implantation", *ELECTROTECHNICA & ELECTRONICA*, 5-6, 2012, 268-272.
- [10] Kakugawa, K., M. Hosotani, M. Arikado, T. Tabe, K. Fukutomi, Y. Tsuchiya, T. Tanaka, "Experimental study on sterilization of food with self-igniting plasma formed from liquid using plasma-based ion implantation", *ELECTROTECHNICA & ELECTRONICA*, 53, 2018, 194-198.
- [11] Morimatsu, K., S. Tanahashi, J. Watanabe, K. Hatou, "A NEW AND RAPID COLORIMETRIC DETERMINATION OF ACETYLCHOLINESTERASE ACTIVITY (in Japanese)", *Eco-Engineering*, 31, 2019, 23-28.