DRY POWDER FOODS STERILIZATION TECHNIQUES BY ATMOSPHERIC-PRESSURE PLASMA

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ABSTRACT Dry powder food ingredients imported to Thailand contain large amounts of viable bacteria and coliform bacteria, and we need a simple, low-cost, dry non-thermal sterilization method without spoiling nutrients, color, fragrance and flavor. In this study, it is shown that the sterilization performance against viable bacteria and coliform bacteria is proportional to the plasma irradiation time when OH radicals are incident on the dry powder food ingredients placed in an atmospheric-pressure non-equilibrium DC pulse discharge Ar + O2 mixture gas plasma jet.

1. INTRODUCTION

The supply of food ingredients including supplements and Chinese herbal medicine greatly depends on both domestic production and importation. The companies taking part in FOSHU (food for specified health use) market are lately experiencing problems – especially food poisoning caused by orally transmitted bacteria – with large amounts of viable bacteria and coliform bacteria in the dry powder food ingredients imported from abroad. As a means to sterilize viable bacteria and coliform bacteria, the facilities for producing FOSHU adopt hot air process or steam process , which causes, due to heat steam, an adverse effect on nutrients, color, fragrance and flavor. To solve this problem, a simple, low-cost, dry non-thermal sterilization method is needed without affecting the qualities such as nutrients, color, fragrance and flavor in the facilities producing FOSHU using dry powder food ingredients containing viable bacteria and coliform bacteria.

To this, the authors have succeeded in introducing the use of the laser-induced fluorescence method – which is often said to be difficult – to observe the OH radicals emitted from oxygen plasma using atmospheric-pressure non-equilibrium discharge plasma. The techniques using OH radicals, attracting attention as sterilization technology, have often been reported in the medical field.

In this study, to sterilize viable bacteria and coliform bacteria without spoiling nutrients, color, fragrance and flavor, a new methodology using an atmospheric-pressure non-equilibrium DC pulse discharge plasma is proposed for the use at low temperatures so as to perform the irradiation of OH radicals to dry powder food ingredients. Thereby, it is shown that the sterilization effect against viable bacteria and coliform bacteria in the dry powder food ingredients increases proportionally to the irradiation time of the plasma irradiated using an atmospheric-pressure non-equilibrium DC pulse discharge Ar + O2 mixture gas plasma jet generated by high frequency pulse power.

2. EXPERIMENTAL SETUP

Fig.1 shows a schematic of the experimental setup. As shown in the figure, plasma was generated using an RF pulse power source for plasma generation (PHF-1K-W manufactured by Haiden Laboratory Inc.) and atmospheric pressure plasma jet electrodes (PJ-6K manufactured by Haiden Laboratory Inc.) with power 0.5 kW and voltage 1.0 kV. The gas mixture of argon (5.0 L/min) and oxygen (1.0 L/min) was used for the plasma. The atmospheric-pressure nonequilibrium discharge plasma jet electrodes can generate dielectric barrier electric discharge over a relatively large area of the upstream part of the torch and efficiently generate active species. In addition, arc discharge is generated in the downstream torch where the dielectric is not present. The system first generates silent discharge using the high-frequency component (rising edge) of a single pulse applied by the RF pulse power source and then generates an arc discharge when the pulse voltage reaches a peak; since the voltage is applied as a pulse wave, the discharge time is as small as several μs and electromagnetic pumping is weak compared with the DC plasma jet. The plasma torch nozzle is made of a titanium rod (4-mm OD × 10-mm length) at the center of body and is covered with a SUS pipe (36-mm OD × 30-mm ID × 87-mm length). A quartz tube (26-mm OD

× 24-mm ID × 87-mm length) is placed between the titanium rod and the SUS pipe at the upper part of plasma torch nozzle. The plasma gas flown into the torch is controlled at 5.0 L/min, and, at the nozzle exit, the speed is increased by means of a small diameter; active species generated by silent discharge in the upstream part of the torch efficiently jet out with the gas force and the electromagnetic pumping action. The heating of the electrodes is prevented and the plasma temperature is kept relatively low because the plasma is supplied by intermittently applying the voltage and the plasma gas flow rate is relatively high .

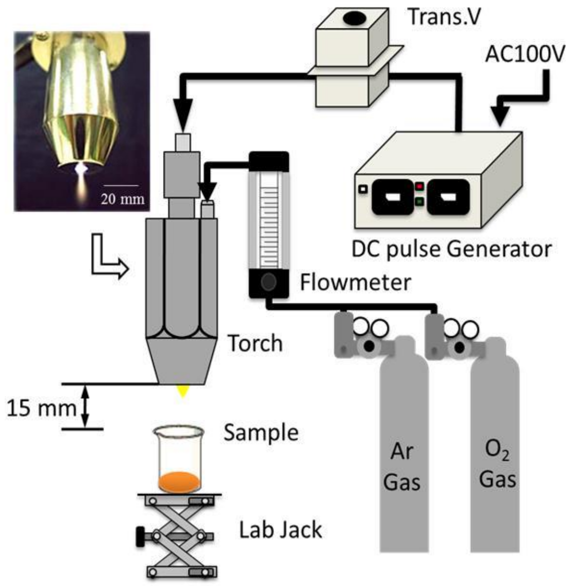


Fig. 1 Schematic of the experimental setup

Figure 2 shows the fine powder of mulberry leaves, which is used as dry powder food ingredients in the experiment. The general bacterial count was 4.2×104 (/g) per gram of the fine powder of mulberry leaves contaminated while the coliform bacteria were of positive types. Generally, the general bacterial count – the count of mesophilic aerobic bacteria such as Salmonella and enterohemorrhagic E. coli O157 , which grow under certain conditions – is generally used as a representative index of the level of microbial food contamination for the comprehensive evaluation of safety, storage stability, and hygienic handling of food. In addition, coliform bacteria are non spore-forming gram-negative bacilli – without specifying the types of bacteria – covering all types of aerobic or facultative anaerobes that break lactose into acid and gas . Note that the fine powder of mulberry leaves, used in dietary supplements such as green juice, contains carotene, calcium, vitamin B1, and iron, rich with protein, minerals, and nutrients such as dietary fibers, and has an effect to improve blood cholesterol levels and neutral fat .

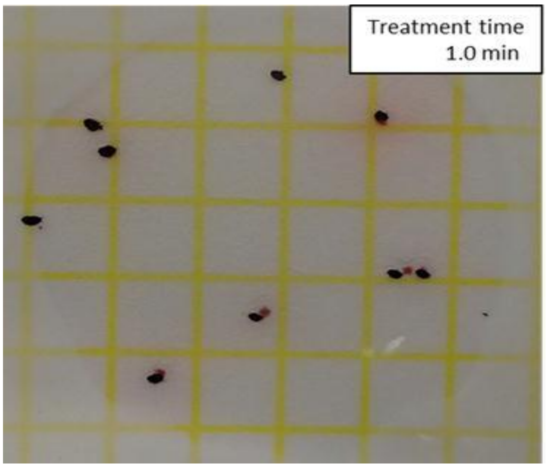
In the experiment, the fine powder of mulberry leaves (20 g) was put in a beaker of 50 mL and placed at the distance of 15 mm from the torch tip so that the fine powder is not scattered by the plasma gas coming from the plasma electrode. The plasma irradiation time was set to one minute, five minutes, and ten minutes, and the fine powder was stirred every two minutes so that the plasma was irradiated on the whole area of the fine powder. The number of general bacteria and coliform bacteria was counted by the PetrifilmTM method with a 100-times diluted solution using PetrifilmTM AC medium for general bacteria and PetrifilmTM CC medium for coliform bacteria.

Fig. 2 Picture of dry food powder (in Beaker)

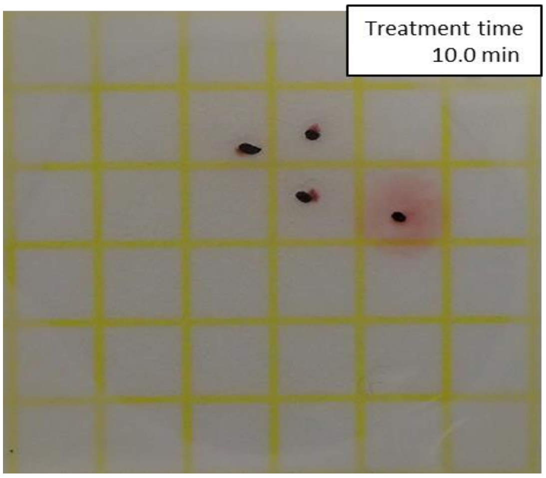
3. RESULTS AND DISCUSSION

Figure 3 shows the distribution of general bacteria per gram of fine powder of mulberry leaves in the PetrifilmTM AC medium evaluated by the PetrifilmTM method in the plasma processing time window. The figure clearly shows that the red dot colonies in the film medium decrease with the plasma irradiation time.

Figure 4 shows the relationship between the general bacterial count per gram of fine powder of mulberry leaves in the PetrifilmTM AC medium and the plasma irradiation time and also the distribution of coliform bacteria per gram of fine powder of mulberry leaves in the PetrifilmTM CC medium over the entire plasma processing time evaluated by the PetrifilmTM method. The figure shows that the general bacterial count is 18×104 (/g) if the plasma processing time is one minute and 8.0×104 (/g) in ten minutes, indicating that half of the bacteria are killed. Furthermore, because coliform bacteria do not exist at all, it is understood that they are negative.



(a) Treatment time 1.0 min



(b) Treatment time 10.0 min

Fig. 3 Picture of AC 3MTM PetrifilmTM in staph express count plate (in Mulberry).

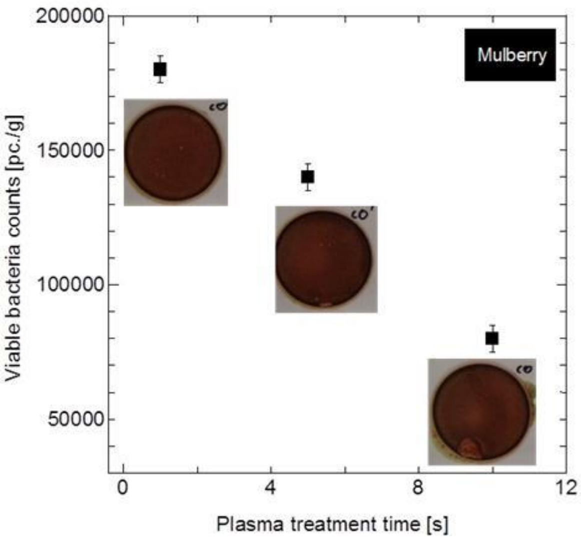


Fig.4 Relationship between plasma treatment time and variety bacteria count and CC 3MTM

In the atmospheric-pressure non-equilibrium discharge plasma processing of the fine powder of mulberry leaves, the general bacteria and coliform bacteria are sterilized because, during the process that the Ar + O2 mixture gas mixture in the atmospheric-pressure non-equilibrium discharge plasma reacts with the electrons emitted from the plasma or the atmospheric moisture, oxygen and water molecules are detached to produce oxygen atoms and, as discussed in earlier reports, ozone and OH radicals are created with a strong oxidizing and sterilizing power. The chemical reaction formulas (1) – (4) are shown below for the creation of ozone in the atmospheric pressure non-equilibrium plasma. where M is any neutral.

$$O_2+e \rightarrow O^{+}_{2}++2e \qquad (1)$$

$$O_2+e \rightarrow 2O+e \qquad (2)$$

$$O_2+O \rightarrow O_3 \qquad (3)$$

$$O+O_2+M \rightarrow O_3+M \qquad (4)$$

Although short-lived, OH radicals with a strong oxidizing and sterilizing power are continually generated as far as ozone and water molecules exist in the atmosphere. We assume that they greatly contribute to the sterilization and inactivation of bacteria. The mechanism of the creation of OH radicals is shown below .

$$e+H_2O \rightarrow e+\cdot OH+H \qquad (5)$$

$$e+O_2 \rightarrow e+O+O \qquad (6)$$

$$e+H_2O \rightarrow \cdot OH+\cdot OH+H \qquad (7)$$

$$O_3+H_2O \rightarrow \cdot OH+\cdot OH+O_2 \qquad (8)$$

4. CONCLUSION

The study using an atmospheric-pressure non-equilibrium DC pulse discharge Ar + O2 mixture gas plasma jet shows that the sterilization effect increases proportionally to the plasma irradiation time, and the general bacterial count in the dry powder food ingredients is halved by the irradiation for ten minutes. The sterilization effect on coliform bacteria was also confirmed. The results suggest that OH radicals with a strong oxidizing and sterilizing power, created by the reaction between plasma radicals and atmospheric moisture, have a great sterilization effect against both general bacteria and coliform bacteria. Ozone and other active species generated in the reacting space play major roles during the degradation process. The discharge process was analyzed. The radical species (O3 and OH, etc.,) were produced in the gas phase and transferred to the air, where they reacted with the substances in it.

In the future, in pursuit of superior performance on dry powder food ingredients, we will examine the plasma gas species and irradiation techniques to complete the dry non-thermal sterilization method using an atmospheric-pressure non-equilibrium DC pulse discharge plasma jet.

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