A DBD-embedded Gas-Liquid Micro Contactor and Its Application to Efficient Depolymerization of Polysaccharides

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Abstract: Gas-liquid micro contactors have been generally employed for realizing efficient gas-liquid interfacial reactions in micro reactors. We have applied this concept for realizing efficient plasma-liquid interfacial processes, in which we employed a DBD-embedded micro contactor (DBD-MC). The DBD-MC is a dielectric plate with multi-hollow DBD electrodes. The liquid-contacting surface of the DBD-MC plate is hydrophobic to prevent the liquid from flowing into the holes (DBD regions) of the plate even if the liquid is in contact with the plate. This system has been applied to the depolymerization of the polysaccharides "Fucoidan" and shown higher energy efficiency than the conventional plasma-in-liquid system.

Keywords: DBD, Micro Contactor, Plasma-Liquid Interface, Depolymerization, Fucoidan

1. Introduction

Plasma-liquid interfacial processes have attracted much attention because they can be used for various applications such as water purification, materials synthesis, medicine, agriculture, and so on [1]. Extensive studies have been made for demonstrating their future possibilities. The most popular plasma-liquid interfacial processes is known as solution plasma processes (SPPs) [2], which employ small plasma generated in a very narrow gap (approx. 1 mm) between two metallic electrodes submerged in a liquid medium as shown in **Fig. 1(a)**. On the other hand, the liquid to be treated is much larger than the size of the plasma and direct effects from plasma do not reach deep into the liquid. Therefore, in order to realize a highly efficient plasma-liquid interfacial processes, it is desirable to make the liquid in contact with the plasma as thin as possible. A wetted-wall plasma reactor is one of the possible solutions [3]. In this case, however, liquid exists as a series component in the discharge circuit and the gas-gap voltage for plasma generation decreases due to voltage drop in the liquid.

To prevent such liquid-conductivity dependence of the sisytem, we have proposed a concept of the DBD-embedded micro contactor (DBD-MC) as shown in **Fig. 1 (b)**, in which liquid can be brought very close without the liquid-conductivity dependence. The liquid-contacting surface of the DBD-MC is hydrophobic, which prevents the liquid from passing through the holes.

In this work, we have applied the DBD-MC for degradation of fucoidan, in other words, for preparation of low-molecular-weight (LMW) fucoidan. Fucoidan is a polysaccharide extracted from seaweeds and characterized by being rich in sulfate groups. When it is ingested in the body, it brings about physiological activities such as induction of apoptosis, anti-blood coagulation, anti-tumor, anti-virus, and anti-allergic effects [4]. The LMW fucoidan (less than 10^3 Da) has much more pronounced physiological activities than as-extracted high-molecular-weight (HMW) fucoidan (10^5–10^6 Da) [5]. For comparison, we have also performed the treatment of fucoidan using a conventional solution plasma processes using the reactor shown in **Fig. 1(a)**.

2. Experimental Procedure

The liquid to be treated with plasma was aqueous solution (1 mg/1 mL) of fucoidan laminaria japonica (BioSynth). Molecular weight distribution of plasma-treated samples was investigated using gel permeation

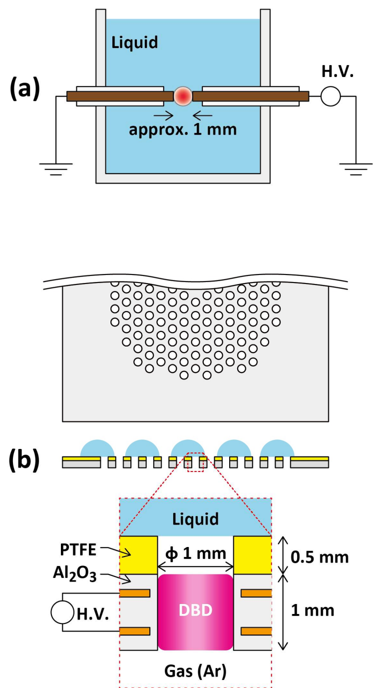

Fig 1. (a) A typical reactor structure for conventional solution plasma processes, and (b) a proposed reactor structure using a DBD-embedded micro contactor.

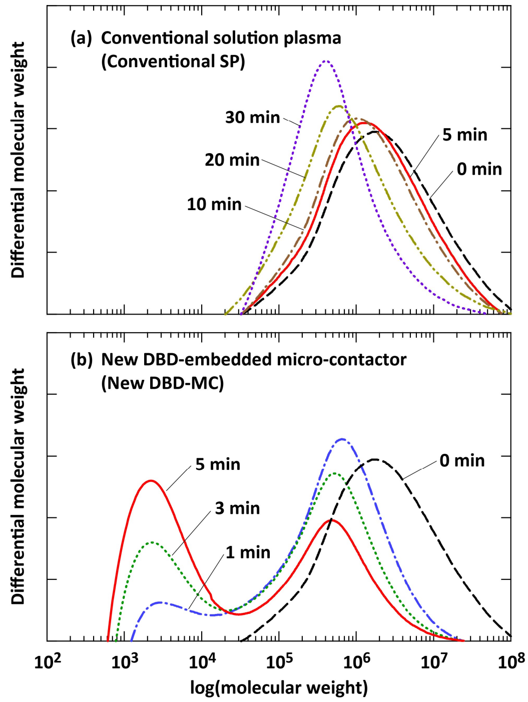

Fig. 2. Molecular-weight distributions of the samples treated with (a) the conventional SPP reactor and (b) the DBD-embedded micro-contactor reactor.

chromatography (GPC) (Shimadzu Nexera), where the column was SHODEX OHpak SB-806M HQ (column temperature: 30ºC, mobile phase: 0.1 M $NaNO_3$, flow rate: 0.4 mL/min). The voltage waveform for the DBD-MC was bipolar and rectangular voltage supplied from semiconductor switches (10 kHz, amplitude 2.7 kV, duty 6% (positve), 6% (negative)). The voltage waveform for the conventional SPP reactor was different from that for DBD-MC due to the limited range of stable plasma generation condition. The voltage waveform for the conventional SPP was bipolar pulses supplied from transformer-boosted inverter (30 kHz, amplitude 400 V, duty 3% (positive), 3% (negative)). The liquid volume for the conventional SPP was 100 mL, while that for the DBD-MC was 7 mL because of the difference in the reactor configurations.

3. Results and Discussion

Figure 2 shows the results of GPC. We can see that the fraction of LMW components in the case of DBD-MC is much more pronounced even after the short treatment time of 5 min while that of conventiona SPP is not. To make a fair comparison, we have compared them under the same energy consumption and the same liquid volume of 100 mL. The consumed energy was calculated as the product of the averaged consumed power and treatment time, where the averaged power was calculated using voltage-current waveforms in the one period. The consumed power for 100-mL treatment in the case of the DBD-MC (7mL per a single reactor) was calculated by multiplying 100/7 to the power for the single reactor.

Table 1. Estimation of the time for 100-mL treatment with the same energy consumption in the cases of the conventional SPP and the DBD-MC.

	Power	Time	Energy
Conv. SPP	12.8 W	10 min	2 Wh
DBD-MC	24 W	5 min	2 Wh

The results are listed in **Table 1**. The consumed power (24 W) of DBD-MC is larger than that (12.8 W) of conventional SPP, which seems to be disadvantage of the DBD-MC. However, if we compare the GPC results under the same energy consumption (2 Wh, for example), we can see that the DBD-MC has superior performance than the conventional SPP. In the case of the conventional SPP, the 2 Wh is consumed in approximately 10 min. The GPC result for the 10-min treatment using the conventional SPP shows slight shift toward lower molecular weight region, but there are no components in the LMW region of 10^2-10^4 Da. On the other hand, in the case of the DBD-MC, the same energy is consumed in 5 min. The GPC result for the 5-min treatment using the DBD-MC markedly different from that for the 0-min treatment and the intensity of the LMW region is much more pronounced than that for the conventional SPP.

4. Conclusion

We have proposed a novel DBD-MC which can be used for plasma-liquid interfacial processes. We applied the DBD-MC to the degradation of fucoidan. Since the DBD-MC handles only thin liquid films, parallel processing is required to process a large amount of liquid, and the power consumption (energy consumption per time) is higher than that of the conventional SPP. However, when compared with the same energy consumption, the amount of decomposition is greater than that of the conventional SPP and required treatment time is much shorter. This is owning to the fact that a thin liquid film can be plasma-treated with very high efficiency when we employ the DBD MC.

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