Corrigendum: The application of a non-thermal plasma generated by gas–liquid gliding arc discharge in sterilization

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The correct version of figure 9 and the caption of the figure are shown here.

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Figure 9. Emission spectrum from gliding arc discharge in air–water.
The application of a non-thermal plasma generated by gas–liquid gliding arc discharge in sterilization

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Abstract. Gliding arc discharge has been investigated in recent years as an innovative physicochemical technique for contaminated water treatment at atmospheric pressure and ambient temperature. In this study we tested a gas–liquid gliding arc discharge reactor, the bacterial suspension of which was treated circularly. When the bacterial suspension was passed through the electrodes and circulated at defined flow rates, almost 100% of the bacteria were killed in less than 3.0 min. Experimental results showed that it is possible to achieve an abatement of 7.0 decimal logarithm units within only 30 s. Circulation flow rates and types of feeding gas caused a certain impact on bacteria inactivation, but the influences are not obvious. So, under the promise of sterilization effect, industrial applications can select their appropriate operating conditions. All inactivation curves presented the same three-phase profile showing an apparent sterilization effect. Analysis of the scanning electron microscope images of bacterial cells supports the speculation that the gas–liquid gliding arc discharge plasma is acting under various mechanisms driven essentially by oxidation and the effect of electric field. These results enhance the possibility of applying gas–liquid gliding arc discharge decontamination systems to disinfect bacterial-contaminated water. Furthermore, correlational research indicates the potential applications of this technology in rapid sterilization of medical devices, spacecraft and food.
New sterilization and disinfection techniques are developing rapidly since hazardous microbes have become a major threat to environmental protection and human health. Among the emerging decontamination techniques, electrical discharges show their advantages for the efficient plasma they produce. Electric discharges are usual sources of plasma gases which are made of mixtures of heavy (i.e. molecules, atoms, radicals and ions) and light species (i.e. electrons and photons) \[1\]. Energy transfer from electric field to gas raises the energy level of some heavy species to an excited state with modified electron distribution and consequently with enhanced or modified reactivity \[2, 3\]. The electrical discharge plasma belongs to the group of non-thermal plasma \[4\] in which the mean energy of the electrons at low pressure is much higher than that of the heavy species, and its key advantages are cheap equipment, flexible operations, high degree of non-equilibrium state and high-efficiency pollutant removal. The gilding arc discharge, a non-thermal plasma technology, has been applied in the inactivation of microorganisms because of its low-cost equipment and high-efficiency decontamination. The principle of the gliding arc discharge was patented by Lesueur and his co-workers in 1988 and then developed by Czernichowski and co-workers \[3, 8\]. It is a simple kind of technique that operates at atmospheric pressure, can adapt well to exploit high electric power and consequently leads to the formation of an abundance of short-lived active species \[9\]. These chemical species are the main particles responsible for a series of complex reactions when they make contact with the outside substance \[10\]. The gliding arc discharge was first investigated for gas treatment \[3, 11\], and decomposition of toluene was also performed and studied. Experimental results indicated that the gliding arc discharge could effectively decompose toluene molecules and had bright prospects of being applied as an alternative tool to decompose volatile organic compounds \[12\].

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Recently, the application of this non-thermal plasma technology was largely devoted to the surface treatment of temperature-sensitive materials.

The gas supply of the gliding arc discharge is generally humid air. As moist air contains a variety of molecular species (N\textsubscript{2}, O\textsubscript{2} and H\textsubscript{2}O), OH· and NO·, the main radicals in the non-thermal phase [13], are formed after the decomposition of those elements under the electric field [14]. The gliding arc discharge has been applied in the inactivation of various microorganisms. In the study by Moreau et al [15], total destruction of Erwinia cultures was obtained in less than 6 min, as well as of various species of the former genus Erwinia [16]. Decontamination of Staphylococcus epidermidis and Hafnia alvei by the gliding arc discharge also appeared to be considerably effective [17, 18]. Disinfection by gas–liquid gliding arc discharge is a novelty motivated by these previous studies. The fact is that O\textsubscript{3}, O· and UV radiations are formed in the gas-phase discharge, while OH· and H\textsubscript{2}O\textsubscript{2} can be formed in the liquid-phase discharge, which makes this technology attractive for various applications [19, 20]. Moreau et al [2] investigated the inactivation of E. carotovora subsp. atroseptica with the gliding arc plasma and achieved better inactivation efficiency than the radio-frequency plasma system in the study by Sharma et al [5]. Niemira et al [6] applied the gliding arc plasma to outbreak strains of Escherichia coli O157:H7 and Salmonella stanley on agar plates and inoculated onto the surfaces of Golden Delicious apples. Their results suggested that gliding arc plasma was a non-thermal process that can effectively reduce human pathogens inoculated onto fresh produce. Kamgang-Youbi et al [7] used the plasma-activated water from a gliding arc reactor to treat planktonic and adherent cells of Staph. epidermidis, Leuconostoc mesenteroides, H. alvei and Saccharomyces cerevisiae and reached a conclusion that the gliding arc plasma-activated water was a promising solution with potential application in the decontamination of equipment and surfaces.

In this study, we decided to investigate the effect of gas–liquid gliding arc on microorganisms and report the results of disinfection of water contaminated by E. coli. This bacterium was chosen for its influence on the water supply, since it is the indicator organism for fecal contamination of water. The survival of E. coli in water treatment and distribution systems has also added urgency to the difficult and heavily regulated process of water disinfection. By far the most common treatment techniques used for the management and reduction of microorganisms in water are UV light irradiation [21, 22] and chlorine [23]. However, the methods have the drawbacks of subsequent pollution problems or residual hazardous substances issues. Gliding arc discharge technology appears to be a promising method complementary to these challenges. We will demonstrate the flexibility and efficiency of gas–liquid gliding arc discharge that allows the rapid and stable disinfection of water contaminated by E. coli. Experimental results are discussed with regard to the potential mechanisms involved in the decontamination process and help us to explore the potential applications of the gliding arc discharge.

2. Materials and methods

2.1. The gas–liquid gliding arc reactor

The experimental setup used in this study consists of an ac high-voltage power supply and a reactor, and derives from the device described in detail in the previous work [24]. The scheme of this apparatus is given in figure 1. It is a non-thermal quenched plasma system operated at atmospheric pressure and ambient temperature. The reactor body is made of a glass cylindrical...
vessel (1 L) with a circulating-water thermostatted jacket to limit the temperature increase during the experiment. The gas and liquid are injected through a nozzle placed in the axis at the top of the reactor. Discharge is produced by two knife-shaped stainless steel electrodes (95 mm long, 35 mm wide and 4 mm thick) working at a high-voltage fall (10 kV, 50 Hz). An arc forms at the electrode minimum gap and is then pushed away by the bi-phase flow from an atomizing nozzle. It ‘glides’ along the electrode walls until it collapses. Once the arc breaks into a plasma plume, a new arc forms at the top of the electrodes and develops in the same way. Thus a consecutive cycle comes into being, and a larger volume of non-equilibrium plasma forms. The bacterial suspension will be supplied with water when it passes through the electrodes with the help of a magnetic pump. Then the bacterial suspension is treated circularly once with a short plasma-exposure time in the plasma zone.

2.2. Bacterial strain and culture conditions

*E. coli* strain ATCC 25922 used throughout this study was purchased from the American Type Culture Collection (Manassas, VA). The strain was maintained as a glycerol stock culture and stored at −80°C until it was used. There also were stock cultures deposited on LB agar slants in the dark at 4°C, which were prepared for inoculation before each experiment. The *E. coli* stock was inoculated into sterilized LB broth (1% tryptone, 0.5% yeast extract and 1% salt) and grown in the dark at 37°C while shaken at 160 rpm. We tested the growth curve of *E. coli* and then used bacteria that were in the late stationary phase to conduct our experiments, since bacteria in the
late stationary phase are particularly resistant to decontamination [25]. The bacterial cells were separated by centrifugation, then washed twice and resuspended in sterilized 1.5 × 10^{-1} M NaCl to obtain bacterial suspensions with initial cell concentration ranging from 10^8 to 10^{10} CFU (colony-forming units) ml^{-1}. For each series of experiments, a 200 ml sample of this suspension was transferred to the reactor and then exposed to the gliding arc discharge. The classic plating technique using LB agar medium in a Petri dish was used to plate culture before and after treatment. A given volume of the sample was implanted on the surface of that plate after a series of gradient dilutions and then incubated for 24 h at 37^\circ C, after which viable colonies could be counted.

2.3. Inactivation efficiency

In this study, the inactivation efficiency by gas–liquid gliding arc discharge is represented by log reduction and the proportion of surviving bacteria. The log reduction was calculated as follows:

\[
\text{log reduction (CFU ml}^{-1}) = \log(\text{the living bacteria concentration of the initial suspension, CFU ml}^{-1}) - \log(\text{the living bacteria concentration of the treated suspension, CFU ml}^{-1}).
\]

The proportion of surviving bacteria (%) indicated a comparison of the living bacteria concentration of the treated suspension to the initial sample that was left untreated. A higher log reduction and a lower proportion of surviving bacteria all represent higher inactivation efficiency. Each experiment was repeated twice to ensure the stability of experimental results. All the data shown in the figures and in the assay were determined over a minimum of three independent media.

2.4. Scanning electron microscopy

In order to prevent destruction after sampling, the E. coli cells were fixed with 2.5% glutaraldehyde for 6 h. Then the cultures were centrifuged (20^\circ C, 5000 g, 10 min) and the relevant supernatant was removed. The corpuscles were rinsed thrice in PBS buffer (0.8% NaCl, 0.02% KCl, 0.17% Na_2HPO_4 and 0.8% KH_2PO_4) and dehydrated by successive treatments of ethanol in water, in which the increase of ethanol concentration for each treatment was a gradient growth (from 30 to 100%). Later the samples were treated with anhydrous tert-butyl alcohol three times and finally dried at the critical temperature. Then all the samples were coated with gold powder in a sputter coater and examined by a scanning electron microscope (SEM) (S-520; Hitachi, Japan).

3. Results and discussions

3.1. Destruction of bacteria by gas–liquid gliding arc discharge

Experimental tests were conducted in the reactor with the bacterial suspension circulated at a defined flow rate, i.e. bacteria were treated by gas–liquid gliding arc discharge (U = 10 kV, \(Q_{air} = 0.8 \text{ m}^3 \text{ h}^{-1}\), \(Q_{liquid} = 20 \text{ ml min}^{-1}\)). Control experiments without discharge were also carried out at the same time. Figure 2 presents the corresponding inactivation efficiency of E. coli, and the effect of gas–liquid gliding arc discharge technique was studied over a period of 3.0 min.

In the case when bacterial suspension is passed through the electrodes, a 2.5 min treatment led to a significant total destruction of the bacterial population. The circulation of bacterial suspension played the role of agitation and made the liquid passing through the electrodes and under the electrodes mix adequately. Samples were mixtures of bacteria passing through the
Figure 2. Inactivation of E. coli exposed to the gliding arc discharge. Operating conditions: $U = 10 \text{kV}$, $Q_{\text{air}} = 0.8 \text{ m}^3 \text{ h}^{-1}$, $Q_{\text{liquid}} = 0 \text{ ml min}^{-1}$, $Q_{\text{liquid}} = 20 \text{ ml min}^{-1}$.

electrodes and beneath the plasma zone. A 200 ml portion of the suspension was placed in the reactor for each experiment, and $Q_{\text{liquid}}$ was 20 ml min$^{-1}$. Only 30% of the mixtures passed through the electrodes directly within three minutes. No cultivable cells could be detected in the samples after 2.5 min, although the last two samples were plated straight without diluting. However, the number of surviving bacteria remained almost steady in 3.0 min and was close to the initial value when there was no discharge in the reactor, which indicated that the mechanical force had little effect on live bacteria. As for the gas–liquid gliding arc discharge treatment, the destruction of the bacterial concentration versus the treatment time presented three phases (figure 2): an initial lag phase, during which the number of surviving bacteria remained relatively high and was almost steady; then the rapid decline phase in which a reduction of 7.0 logarithm units of the bacterial population was obtained within 30 s; and the last phase was a stable state in which no cultivable cells could be detected on the Petri dishes.

These significant sterilizing results of the gas–liquid gliding arc discharge with air as a carrier gas can be attributed to several factors described below. Firstly, the enormous energy involved in the discharge plays a major role when the bacterial suspension passes through the electrodes. A high electric field of 10 kV cm$^{-1}$ during the formation of electrical discharge can effectively inactivate any microbes [9], crush the microbial cells or even burn them to carbon. When the suspension passed through the electrodes, the bacteria within were exposed to a high temperature (5000 K) in the thermal zone, which can easily cause a lethal effect, although there is a thermostatic circulating bath to control the temperature of the liquid below the electrodes between 24.9 $\pm$ 0.2 and 27.4 $\pm$ 0.2 $^\circ$C. Whereas at most 30% of the suspensions passed through the electrodes directly within 3 min, more than 70% of the bacteria were treated by the non-thermal plasma. Hence, the non-thermal effect of the reactive species is the major bactericidal
effect. An important factor is the physical and chemical effects related to the formation of some radicals in the gliding arc discharge plasma. In the physical effects that always accompany the discharge, UV radiation is thought to be an important factor in sterilization. Using air as a carrier gas, and if water exists, the chemical active species formed in the gliding arc discharge include short-lived active species (·OH, NO·, O·, O2·−, HO2· and H·) and long-lived active molecules (H2O2, O3 and other excited-state neutral molecules); related reactions are as follows [26]:

\[
\begin{align*}
H_2O + e^- & \rightarrow H\cdot + \cdot OH + e^- \\
O_2 + e^- & \rightarrow O (^3P) + O (^1D) + e^- \\
N_2 + e^- & \rightarrow N (^4S) + N (^2D) + e^- \\
N (^2D) + O_2 & \rightarrow NO\cdot + O \\
NO\cdot + \cdot OH & \rightarrow HNO_2 \\
H\cdot + O_2 & \rightarrow HO_2\cdot \\
HO_2\cdot + HO_2\cdot & \rightarrow H_2O_2 + O_2 \\
HO_2\cdot + NO\cdot & \rightarrow NO_2\cdot OH \\
\cdot OH + \cdot OH & \rightarrow H_2O_2 \\
O_2 + O (^3P) & \rightarrow O_3
\end{align*}
\]

The circulated bacterial suspension provides us with sufficient water; then ·OH and NO· are the main species formed in humid air plasma during the discharge [27, 28]. NO· is responsible for acid effects and ·OH is the oxidizer associated with H2O2; the two effectively contribute to the inactivation of bacteria together with all the above actions.

3.2. Sterilization effects with air under different circulation flow rates

When air was used as a carrier gas, the proportions of surviving bacteria in the suspensions treated by gas–liquid gliding arc discharge under different circulation flow rates had similar decrease trends. The destruction curves of E. coli presented in figure 3 all show similar three phases as described above, in which a complete inactivation of the bacterial population could be achieved in 2.5 min. There was no significant difference between the inactivation efficiencies of E. coli under the four flow rates, except that a higher flow rate had a shorter lag phase and the falling trend of surviving bacteria concentration appeared slightly obvious. While the flow rate reached 50 ml min⁻¹, the time required to complete the whole treatment was shortened to only 2.0 min. However, energy consumption should be considered and industrial applications can select an appropriate flow rate corresponding to operating conditions.

Analyzing the slight influence of the circulation flow rate, we can easily find that the increase of flow rate will do some favor to the inactivation of E. coli suspended in water. Suspension circulated at a higher flow rate makes more bacteria per unit pass through the discharge electrodes, which at the same time means more microbes will be inactivated instantly. Higher flow rates of the circulating liquid can also result in a shorter time of flight from the region of active plasma to the target suspension. On the other hand, the growth of the cycling rate.
Figure 3. Sterilization effects under different circulation flow rates with air as the carrier gas. Operating conditions: \( U = 10 \text{kV} \), \( Q_{\text{air}} = 0.8 \text{ m}^3 \text{ h}^{-1} \).

accelerates the mixture of the discharge products and bacterial solution; then the free radicals and active substances can fully contact and react with the microorganisms. It is worth noting that a flow rate of 20 ml min\(^{-1}\) can already achieve a pretty good sterilization effect, so higher flow rates need not be selected since the enhancements are not obvious.

3.3. Sterilization effects with nitrogen under different circulation flow rates

The effect of the gas–liquid gliding arc decontamination technique on the bacterial cells of \( E. \text{coli} \) was also studied with nitrogen as a carrier gas. In this case, the bacterial suspension was circulated at different flow rates and the treatment time was equal to the discharge using air as the feeding gas. Correspondingly, the inactivation efficiencies are shown in figure 4. The destruction curves and the fallen trends are similar to the experiments using air, including the influence of the circulation flow rate. After treatment for 3 min, a total destruction of the \( E. \text{coli} \) population could be achieved and the variation of numbers of surviving bacteria presented a familiar pattern described in former tests. Like the air–liquid gliding arc discharge, a higher flow rate could slightly shorten the treatment time required for total inactivation of the microorganisms.

It was shown that the gliding arc technique using nitrogen as the gas resource could effectively inactivate bacteria in flowing water in a very short time. The huge energy of electric field, high temperature between electrodes, shockwave and chemicals that are formed in the discharge all contribute to the microbial inactivation. As for the formation of chemical active species during the discharge, the related reactions are somewhat different from the conditions with air. Since there is just nitrogen in the feeding gas, \( \cdot \text{OH} \) and \( \text{H}_2\text{O}_2 \) are produced by reactions (1) and (9) and little NO- or \( \text{O}_3 \) can be detected in the discharge [29]. Differences in inactivation efficiency between the two feeding gases will be explained in the following discussions.
Figure 4. Sterilization effects under different circulation flow rates with nitrogen as the carrier gas. Operating conditions: \( U = 10 \text{kV}, Q_{\text{nitrogen}} = 0.8 \text{ m}^3 \text{h}^{-1} \).

Figure 5. Sterilization effects under different circulation flow rates with oxygen as the carrier gas. Operating conditions: \( U = 10 \text{kV}, Q_{\text{oxygen}} = 0.8 \text{ m}^3 \text{h}^{-1} \).

3.4. Sterilization effects with oxygen under different circulation flow rates

Experimental results obtained with E. coli suspension circulated at different flow rates when oxygen was used as the feeding gas are shown in figure 5. The destruction was slightly more efficient than the treatments that employed air and nitrogen as carrier gases, although the
profile in figure 6 appears similar to the two described above. From the figure, we notice that the length of the lag phase is slightly shorter than previous curves, and the decay of the cultivable population seems steeper. These variations reveal that when oxygen was employed, the inactivation reactions started faster and more microorganisms were sterilized in a very short period. When the flow rates reached 40 and 50 ml min$^{-1}$, no cultivable cells could be detected on the Petri dishes after 2 min treatment and complete inactivation of microorganisms in liquid was realized.

The products of the gliding arc discharge with pure oxygen are not exactly the same as the conditions with air and nitrogen. Adequate oxygen promotes the plentiful formation of O$_3$ by reactions (2) and (10), which is a powerful oxidizing agent and can readily form reactive ·OH and HOO· radicals in aqueous solution [30]. Meanwhile, a considerable number of H$_2$O$_2$ is formed in the suspension through reactions (1), (6), (7) and (9). All these discharge products, together with the physical effects of electric field, high temperature and shockwave during the discharge, interact with the microorganisms and result in a significant bactericidal effect.

3.5. Sterilization effects with argon under different circulation flow rates

The decontamination performance of the gliding arc treatment on E. coli with argon as a feeding gas has the same features as those reported with other gases. Figure 6 shows the results when the bacterial suspension was exposed to discharge, in which the influence of circulation flow rates is as usual within the expected range. The inactivation efficiency of this case is not illustrated due to the similarity of the inactivation efficiency among these four carrier gases. Using argon as the feeding gas has a disinfection effect that is even better than the conditions with oxygen. The experimental results revealed that gas–liquid gliding arc discharge plasma treatment with higher circulation flow rates not only rapidly reduced the bacterial population, but also shortened the total sterilization time. When the flow rates reached 30, 40 and 50 ml min$^{-1}$, the numbers of
detected surviving cells all fall to zero after 2 min of treatment although argon is not frequently used under common conditions.

There is no nitrogen or oxygen in the discharge when the carrier gas is pure argon. Chemical mixtures during the discharge are formed by reactions (1) and (9) and the other three listed below:

\[ \text{Ar} + e^- \rightarrow \text{Ar} + 2e^- \]  
\[ \text{Ar} + \text{H}_2\text{O} \rightarrow \text{Ar}^+ (\text{H}_2\text{O}) \]  
\[ \text{Ar}^+ (\text{H}_2\text{O}) + \text{H}_2\text{O} \rightarrow \text{H}_3\text{O}^+ + \text{Ar} + \cdot \text{OH} \]

In addition to the physical effects generated during the discharge, another possible sterilization mechanism of gliding arc discharge plasma with argon at atmospheric pressure is the continuous impact on the membrane of bacterial cells of energetic plasma species such as argon ions, metastable particles, electronically excited neutrals, together with the corresponding derivatives. It is said that H\(_2\) and H\(_2\)O\(_2\) production in a gliding arc reactor with water spray largely depends on the nature of the carrier gas, with the highest production rate found with argon carrier gas [31]. The characteristic energies of argon neutrals (including argon metastables and electronically excited argon atoms) and argon ions are of the order of 15 and > 11 eV, respectively, which are much higher than the bonding energy of the organic molecules constituting microorganisms [32]. That means that only pure argon plasmas have the ability to destroy various microorganisms. This unique feature combined with the effects and discharge products that exist in other gas plasmas creates remarkable bacterial inactivation efficiency.

### 3.6. Comparison of bactericidal efficiencies with different carrier gases

Since the feeding gas is an essential factor in plasma treatment, comparative experiments among different gases were conducted and the results are shown in figure 7. Experiments were carried out with the *E. coli* suspension circulated at a defined flow rate (20 ml min\(^{-1}\)), and all the gases were injected into the reactor at the same optimum gas velocity. Figure 7 shows the inactivation curves for *E. coli* planktonic cells subjected to gas–liquid gliding arc discharge, and all four curves present similar kinds of changes. Inactivation of *E. coli* followed a time-dependent reduction for all carrier gases. For all four kinds of treatment, a successive discharge of 3.0 min was enough to kill all the bacteria in the suspension; meanwhile, a reduction of not less than 7 logarithm units of the bacterial population was obtained within 30 s. The results in figure 7 also show that the choice of the carrier gas plays a very limited role in the promotion of inactivation efficiency, since the four curves have common profiles and the intervals are relatively small. However, we can also find that the disinfection effects of gas–liquid gliding arc discharge treatment with air, nitrogen, oxygen and argon appear to be quite similar. Experimental results also show that argon has the best inactivation effect when it is employed as a feeding gas.

Gas–liquid gliding arc discharge treatment with air as a feeding gas already has a rather remarkable disinfection effect described above. The change of carrier gas type affects the formation of several active species (·OH, H\(_2\)O\(_2\) and O\(_3\)) and byproducts such as NO\(_2^-\) and NO\(_3^-\). Emission spectrum measurements [29] revealed that the relative intensity of ·OH hydroxyl radical is highest for air as the feeding gas, in turn followed by argon, oxygen and nitrogen.

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The $\text{H}_2\text{O}_2$ concentration in the case of oxygen is higher than that of air, nitrogen and argon. Ozone is produced when air and oxygen are employed; however, there is no $\text{O}_3$ in the case of nitrogen and argon. The decrease of pH, which is accompanied by an increase of conductivity, can partially be explained by the formation of $\text{H}_3\text{O}^+$ irons in the water. Simultaneously, the $\text{NO}_2^-$ and $\text{NO}_3^-$ formed in the water are in part accountable for the pH decrease and conductivity increase with nitrogenous gas. All these complex reactions and transformations, in conjunction with the physical effects during the gliding arc discharge, contribute to the connections and slight differences of inactivation efficiency among the four gases. It is necessary to put forward that power consumption in the study is related to the flow rate and the feeding gases. Although the increase of flow rates and the selection of some gases can improve the bactericidal effect, the promotion level is not significant. Therefore, gas–liquid gliding arc discharge at a reasonable flow rate with air as the carrier gas is recommended for practical applications.

3.7. SEM images of plasma-treated cells

SEM observations of the bacteria submitted to gas–liquid gliding arc discharge visually illustrate the effects on bacteria of the discharge plasma. Samples were taken from the experiment with air as the carrier gas, and the circulation rate is 20 ml min$^{-1}$. Figure 8(A) presents the $E. \text{coli}$ samples before discharge treatment, in which the cell membrane is smooth and the morphology is fairly complete. However, it can be seen that $E. \text{coli}$ cells are severely damaged and there remains much cell debris in the gas–liquid gliding arc discharge plasma sterilized samples (figure 8(B)), which indicates that the overall structure of the microorganism is significantly affected.
Figure 8. SEM images before and after gas–liquid gliding arc discharge treatment. (A) E. coli cells before plasma treatment. (B) E. coli cells after the whole process of plasma treatment.

3.8. Mechanism of bacteria inactivation

A great deal of damage can be done to bio-macromolecules by gas–liquid gliding arc discharge, but the dominating damage that leads to death is not always very clear. We studied the mechanism of bacteria inactivation through experiment under defined conditions with air as a carrier gas. UV radiation has been proposed as the main component of bactericidal action during the discharge, since it is related to the irreparable damage of DNA and RNA [33]. Biological impact of UV radiation is primarily due to the absorption of photons by nucleic acids. In bacteria, the various repair mechanisms are generally quite efficient and rapid [30], which means that cell death occurs only when the UV photon hits are sufficiently plentiful that the bacterial repair mechanisms cannot manage [34]. However, the number of UV photons generated in the discharge is limited [16], so the corresponding lethal doses are not delivered and the sterilization effect observed with gliding arc discharge probably involves other factors. Ozone is a powerful oxidizing agent, which is formed by reactions (2) and (9) when the carrier gas contains oxygen. The gaseous ozone can react with the fatty acids and peptides of the cell wall [35], stimulate the lipid peroxidation [36] and result in single-strand DNA breaks [37], which eventually lead to extensive DNA damage and death. Bacterial inactivation by ozone is a function of ozone concentration per viable bacterium. However, water vapor in the gas–liquid medium can absorb a substantial part of the electronic energy of the discharge that could otherwise be used in the ozone formation process [29], which indicates that the production of gaseous ozone is below the threshold concentration ozone that can inactivate bacteria. Consequently, the chemical processes of oxidation and acidification occurring during the discharge are proposed to be the main mechanisms to explain the inactivation of bacteria. A spectroscopic investigation of the gas–liquid gliding arc discharge revealed that the radicals NO$^\cdot$ and -OH were the main species formed in the non-thermal phase (figure 9). Moreau et al [16] demonstrated that NO$^\cdot$ is responsible for acid effects and for a steep lowering of the pH of the medium, since it can lead to the formation of HNO$_2$ and HNO$_3$. However, the variation
of pH detected in the 3.0 min treatment is inconspicuous (only changes from 6.84 to 5.78), meaning that the acid effects are not obvious in the sterilization process. The other radical ·OH, which is produced in high quantities, is responsible for a strong oxidizing effect [13, 28], having extremely high rate constants for reactions with almost every type of molecule found in living cells [38] and causing significant damage to most biological molecules. Additionally, the \( \text{H}_2\text{O}_2 \) formed through reactions (1), (6), (7) and (9) in the aqueous medium is also an oxidizing agent that has very high sterilization activity, which can damage the DNA and result in the death, mutation or carcinogenesis of cells [39]. Furthermore, the enormous energy of the high electric field during the discharge cannot be ignored. Any bacteria that pass through the electrodes will be easily crushed into pieces or even burnt to carbon, and the SEM images presented above can confirm this demonstration. Thus come the synthetic effects and the correlative possible mechanism in the gas–liquid gliding arc discharge treatment. This technology can lead to total destruction of the bacterial population, even when the initial concentration of the bacteria suspension reaches \( 10^{12} \text{ CFU ml}^{-1} \) [15]. A gas–liquid gliding arc discharge has an even better efficiency. Inactivation of microorganisms that are attached to the material surface [40–43] simultaneously reminds us of a direction for the potential application of gliding arc discharge plasma.

4. Conclusion

This study involves the use of a gas gliding arc reactor (the control experimental setup) and a gas–liquid gliding arc discharge reactor with circulation. For a reactor without discharge and the suspension circulated, almost no sterilization effect could be observed. For a reactor with gliding arc discharge and bacterial suspension circulated at a defined rate, all the microorganisms are destroyed in the liquid rapidly, i.e. the gas–liquid discharge reactor has a remarkable effect on bacteria killing. Different circulation flow rates and feeding gases will have a certain impact.
on bacteria inactivation; however, the contributions are not obvious. In all cases, the destruction curves of *E. coli* all present the same three-phase profile. SEM images of the bacteria before and after treatment are consistent with the analysis of inactivation mechanisms, which found that gas–liquid gliding arc discharge plasma acts under various mechanisms driven essentially by oxidation and the electric effect. Altogether, these results allow the development of gas–liquid gliding arc decontamination systems at an industrial level, especially the sterilization of mobile liquid with a suitable flow rate using air as the feeding gas. What is more, the H$_2$O$_2$ formed through those reactions can play the same role as chlorine dioxide without any side effects. The flexibility and efficiency of gas–liquid gliding arc discharge are not only beneficial for the inactivation of hazardous microbes in water but also can be considered for application in particular fields such as sterilization of medical devices, spacecraft and food for avoiding thermal effects or damaging the original properties of the materials.

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