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A facile modification method of activated carbon by spark discharge of atmospheric pressure plasma jets to improve its adsorption performance of methylene blue

Chengzhu Shen <sup>a, b</sup>, Guolin Song <sup>a</sup>, Guoyi Tang <sup>a, b\*</sup>

<sup>a</sup> Advanced Materials Institute, Graduate School at Shenzhen, Tsinghua University, Shenzhen 518055, China

<sup>b</sup> School of Materials Science and Engineering, Tsinghua University, Haidian District, Beijing 100084, China

\* Corresponding author. Tel./fax: +86 75526036752

E-mail addresses: tanggy@sz.tsinghua.edu.cn (G. Tang).

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## Abstract

In this study, spark discharge of atmospheric pressure plasma jets (SDAPPJs) was employed to modify activated carbon (AC) prepared from coconut shells. The physical and chemical properties of AC were tailored by SDAPPJs treatment to enhance the adsorption performance. The pristine AC and its modified samples were characterized by N<sub>2</sub> isotherms, X-ray photoelectron spectroscopy (XPS), fourier transform infrared spectroscopy (FT-IR) and scanning electron microscopy (SEM) in terms of micro structure, chemical composition, morphology, and etc. The methylene blue (MB) removal capability of these AC samples from aqueous solution was investigated by UV-vis spectrophotometer to determine the influences of the modification on the adsorption performance. The results show that

SDAPPJs modification not only improves the specific surface area and total pore volume of AC while maintaining good structural stability, but also increases its surface oxygen-containing functional groups under the influence of active species, which eventually enhances the electrostatic interaction between contaminants and defective surface layers of AC. In brief, SDAPPJs treatment has a positive impact on the adsorption capacity of AC, and its removal efficiency of MB increases by 15.64% when the treatment distance, speed and time are 20 mm, 10 mm/s and 3 min, respectively.

Keywords: Atmospheric pressure plasma jets; Spark discharge; Activated carbon; Adsorption performance; Specific surface area

## 1. Introduction

With low-cost, a wide range of applications and environment-friendly features, activated carbon (AC) is commonly used as an adsorbent and it has been recommended to attain stringent, permissible limits for heavy metal ions and organic pollutants[1, 2]. Especially, it has large specific surface area, controllable pore structure and versatile functional groups on the surface, which bring a broad range of application prospects in air purification, sewage treatment[3-5] and energy storage[6, 7]. However, the properties of AC still require to be improved through modifications on account of its unsatisfactory adsorption performance in practice.

The adsorption performance of AC strongly depends on its specific surface area, total pore volume and pore size distribution[8]. Moreover, for the adsorption of polar molecules, such as methylene blue (MB) or methyl orange (MO), it can be strengthened by electrostatic or dispersion interactions with the oxygen-containing groups or  $\pi$ - $\pi$  coupling electrons present on the defective carbon surface[9-11]. The adsorption capacity of AC is expected to be improved since the more ideal pore structure, specific

surface area and surface functional groups can be obtained through surface modification methods[4, 12].

In previous studies, the surface physical and chemical properties of AC, which play a key role in its adsorption performance, were often enhanced via physical or chemical modification methods[10]. Particularly, some commonly used modification methods including acid[10, 13], alkali[14], as well as heating modification[15] would change both physical properties (total pore volume, pore size distribution and specific surface area) and its chemical properties, such as the types or quantities of surface functional groups[16, 17]. However, these modification methods are often accompanied by complex processes, serious pollution and high cost.

With respect to atmospheric pressure plasma jets (APPJs), it can generate high concentrations of active species without vacuum systems at low temperatures[18]. Usually, these species are composed of a large number of ions, electrons, free radicals and so on. APPJs have been frequently applied in material surface modification (the primary step of functionalization) because the active substances would break the surface chemical bonds of most substances[19]. Notably, if the gas source of plasma varies, different types of functional groups could be formed on the material surface[20]. In addition, the treatment has attracted more interests due to its apparent advantages of low energy consumption, less pollution, controllable modification process and high efficiency[21]. Moreover, when the APPJs was positioned far from AC, only chemical properties might be improved[5, 22]. However, if the distance shortened, spark discharge (a kind of high-energy spark channels) would be generated at the tip of APPJs, as shown in Fig. 1, which could significantly change both surface textural and chemical properties of AC. Although a number of papers have reported APPJs could change the properties of

AC[2, 23, 24], spark discharge of atmospheric pressure plasma jets (SDAPPJs) modification was rarely studied. In this work, the influences of SDAPPJs treatment on AC properties were systematically investigated.



Fig. 1. The phenomenon of SDAPPJs.

## 2. Experimental

### 2.1. Materials

All chemical reagents used in the current study were supplied by Aladdin (Shanghai, China). Methylene blue (MB) is analytically pure and potassium bromide (KBr) is spectroscopically pure. All solutions were prepared with high purity water. Raw materials used in the experiment were commercial AC produced by Xingwanbang Activated Carbon Co., Ltd (Shenzhen, China), and they were prepared from coconut shells (CS) via the phosphoric acid activation.

### 2.2. Surface modification of AC by SDAPPJs

Prior to the experiment, AC samples were pretreated at 105 °C for 10 hours in order to remove adsorbed water and volatile contaminants. The weight of AC treated was about 20 g each time. The APPJ equipment used in the process was PZ2 (Relyon-Plasma, Germany), the input gas was air and main configurations of the equipment are listed in Table 1.

Table 1. Main configurations of APPJs used to modify AC.

Working	Counting	Positive	Negative	Ozone
voltage (V)	distance (cm)	Ions (million/cm <sup>3</sup> )	ions (million/cm <sup>3</sup> )	concentration (ppm)
220	20	11.46	-9.9	0.232

The working distance of the plasma equipment is adjustable according to the processing requirements. However, it must be precisely controlled to facilitate the occurrence of SDAPPJs. Experimental parameters for these samples are listed in Table 2.

Table 2. Experimental parameters for different samples in the SDAPPJs modification process.

Sample code	V (v)	Power (w)	T (min)	Distance (mm)	Speed (mm/s)
AC-0	220	30	0	20	10
AC-1	220	30	3	20	10
AC-2	220	30	5	20	10
AC-3	220	30	7	20	10
AC-4	220	30	10	20	10

### 2.3. Characterization

The surface morphology of AC with and without SDAPPJs treatment was studied by using a scanning electron microscopy (SEM, scanning microscope type S-4800, HITACHI, Japan). During the SEM observations, the typical values of voltage and working distance of the equipment were 5 kV and 9 mm, respectively. Each sample was observed twice, the first observation was to obtain the pristine morphology and its precise position was then marked. When SDAPPJs treatment ended, the second scanning was carried out on the marked area of the sample to acquire the in-situ modified morphology.

The N<sub>2</sub> adsorption isotherms at 77 K was evaluated using an automatic gas adsorption apparatus (BelSorp Max 2013). The BET method was used to calculate the specific surface area ( $S_{\text{BET}}$ )[25], and total pore volume ( $V_{\text{total}}$ ) was obtained by the volume of nitrogen at a relative pressure of ~0.995. Moreover, mean pore diameter ( $D_{\text{mean}}$ ) could be estimated from the Dubinin-Radushkevich (D-R) plot through N<sub>2</sub> adsorption[26].

The variances of the oxygen contents and functional groups on the surface of AC samples were identified by Fourier transform infrared spectroscopy (FT-IR) and x-ray photoelectron spectroscopy (XPS). FT-IR analyses were carried out on a Bruker VERTEX 70 spectrophotometer. The obtained spectra were in the range of  $500\text{ cm}^{-1} - 4000\text{ cm}^{-1}$ . Due to the high light absorbance of AC, a very thin KBr pellet was prepared for each sample with a mass ratio of 500:1 (KBr: AC)[10]. For XPS analyses, the spectra were acquired with a Kratos AXIS Ultra DLD, adopting a monochromatic Al K $\alpha$  x-ray source.

#### 2.4. Adsorption performance evaluations of AC

The adsorption performance of pristine and modified AC was evaluated by UV–vis analysis technique (Scinco S-4100 spectrophotometer) in this study. MB is a typical dye and often used to evaluate the adsorption performance of AC. The initial concentration of MB is 1.6 mg/mL in the tests. Since AC is a type of amphoteric material which possesses both basic and acidic functional groups on the surface[10], so its surface can be positively or negatively charged depending on the solution pH condition. Hence, neutral solutions were needed because of the adsorption performance was susceptible to the positive or negative ions on the AC surface[10]. The volume of MB solution and the mass of AC were 50 ml and 0.1 g, respectively. The adsorption process was conducted in an ultrasonic bath with a constant frequency of 35 KHz at 30°C for 120 min. Then, the solid particles were separated from the mixed solution through a centrifuge (8000 rpm for 8 min). Finally, the concentration of MB remained in the solution was determined by its absorption band at ~664 nm in the spectrum.

### 3. Results and discussion

#### 3.1. Textural properties of AC

##### 3.1.1. Specific surface area and total pore volume

The AC samples modified by SDAPPJs for different time durations were characterized in terms of specific area and total pore volume to determine the changes of textural properties. Following IUPAC classification, the N<sub>2</sub> adsorption-desorption isotherms of pristine AC and SDAPPJs-treated AC were deemed to be type I (Fig. 2.a). Through N<sub>2</sub> adsorption-desorption isotherms and corresponding calculations,  $S_{\text{BET}}$ ,  $V_{\text{total}}$  and  $D_{\text{mean}}$  of these samples (AC-0, AC-1, AC-2, AC-3 and AC-4) were obtained. As shown in Table 3, the specific surface area of AC increases with the treatment time during the first 3 min, while it gradually fall down when the SDAPPJs treatment time prolonged, the trend is



similar to the AC modification study[28]. Specifically, when the AC is treated by SDAPPJs for 3 min, its specific surface area and the total pore volume improves by 9.71% and 9.13%, respectively. The possible reason is that SDAPPJs produced a large number of fresh pores and make the closed pores and dead end pores partly transformed into open pores or through pores within the treated area of AC samples (Fig. 2.b). After 3 min treatment, the specific surface area of AC sample gradually decreases with the extension of the processing time. When SDAPPJs treatment time extends to 10 min, the specific surface area of the sample only increases by 2.01% compared to its pristine state. Such a phenomenon is due to the partial damages of AC micropores by the excessive energy input from APPJs[28]. In fact, the intricate porous structure of AC determines that the specific surface area is predominantly obtained by internal micropores[29]. Therefore, the fluctuation of the surface area is similar to the changes of the total pore volume, and the total pore volume increases by 9.13% when the specific surface area is maximal. However, the mean pore diameter almost remains intact in the whole modification process (Table 3).

Table 3. Textural properties of AC through various modification time.

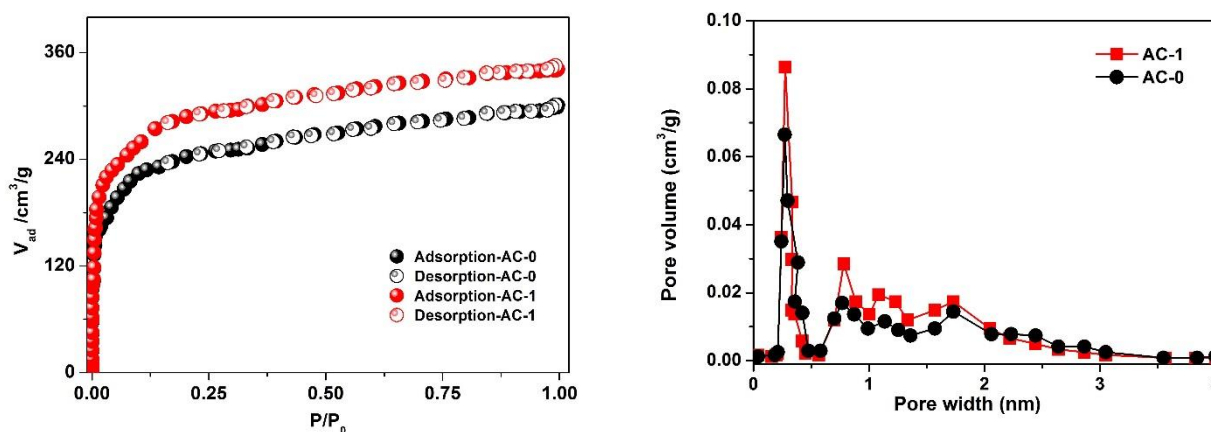


Fig. 2.  $N_2$  adsorption-desorption isotherms (a), and pore size distribution (b) of untreated Samples  $V_{total} (\text{cm}^3/\text{g})$   $S_{BET} (\text{m}^2/\text{g})$   $D_{mean} (\text{nm})$  and SDAPPJs-treated AC.

AC-0	0.3887	954.64	1.6285
AC-1	0.4242	1047.6	1.6197
AC-2	0.4092	1013.4	1.615
AC-3	0.3971	981.46	1.6186
AC-4	0.3911	973.82	1.6065

### 3.1.2. SEM analysis

SEM analysis results of AC samples are presented in Fig. 3. SEM images clearly show that SDAPPJs treatment has a positive impact on AC surface morphology. When SDAPPJs occurs, it emits certain amount of energy through numerous spark discharge channels, which turn out to be streams of small high-velocity plasma packets/bullets instead of continuous plasmas for the conducting materials[30]. In this case, the conductive AC is equivalent to the grounded electrode. In practice, various types of in-situ pores can be generated when these bullets bombard the surface of AC samples.

The increased quantities of pores on the surface enlarge the specific surface area because it would produce fresh pores and make inner closed channels of AC turn into open or through ones, which has been verified by the N<sub>2</sub> adsorption isotherm results. SEM images recorded the in-situ surface morphology variances, which provides a solid proof that SDAPPJs could be an efficient way to improve the surface structure of AC.

In fact, the majority of pores within AC belong to micro-meso level, but the surface pores generated by SDAPPJs are macro-level. In other words, the technique may cause some undesirable damages to the material structure under inappropriate parameters. Briefly, it is a competitive relationship between opening up the internal closed channels and the damage of micro-meso pore structure to certain level.

It gives a reasonable explanation to the phenomenon that the specific surface area does not increase gradually but decline with the extension of SDAPPJs treatment time.

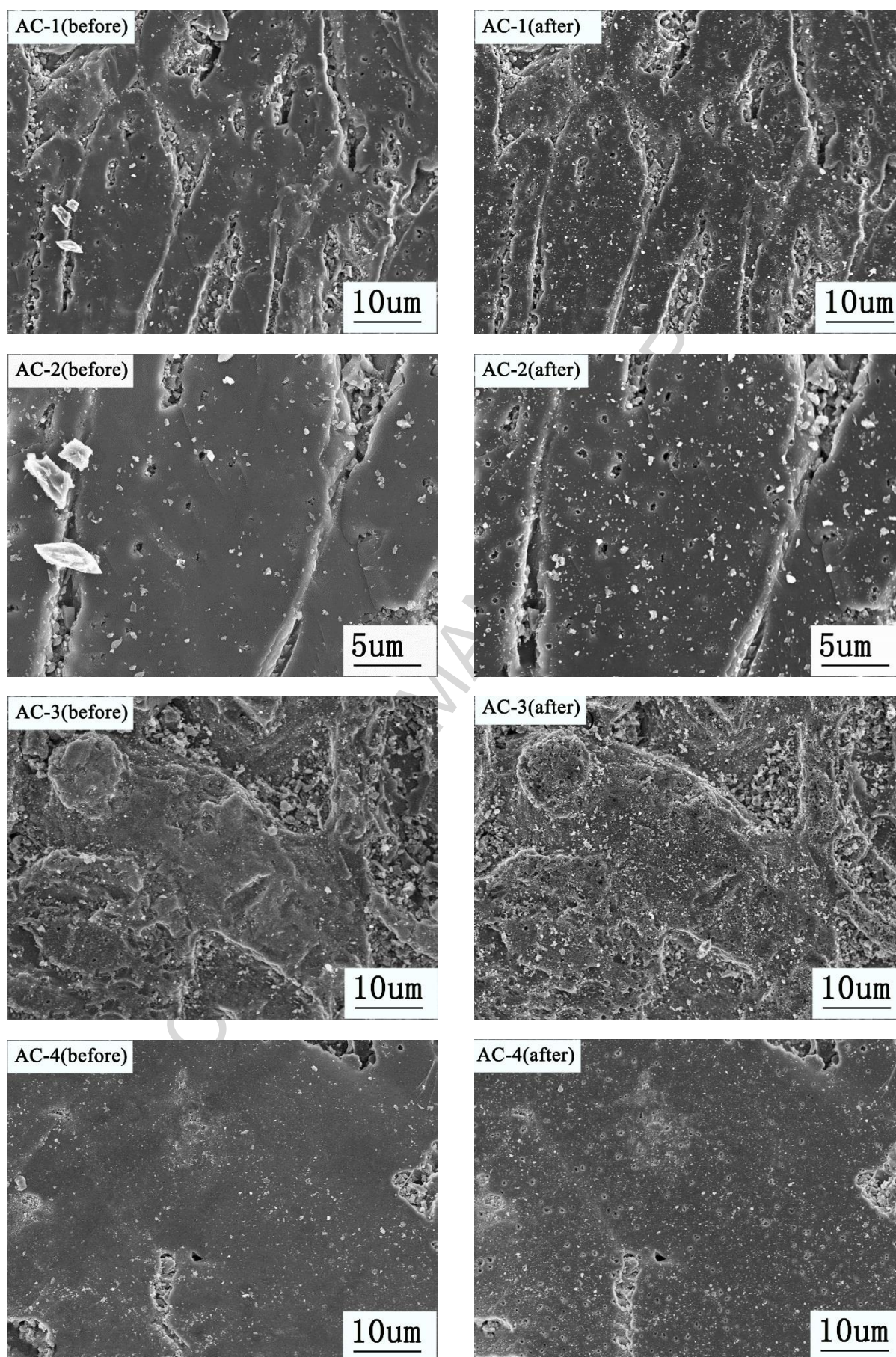


Fig. 3. In-situ SEM images of AC before and after SDAPPJs treatment.



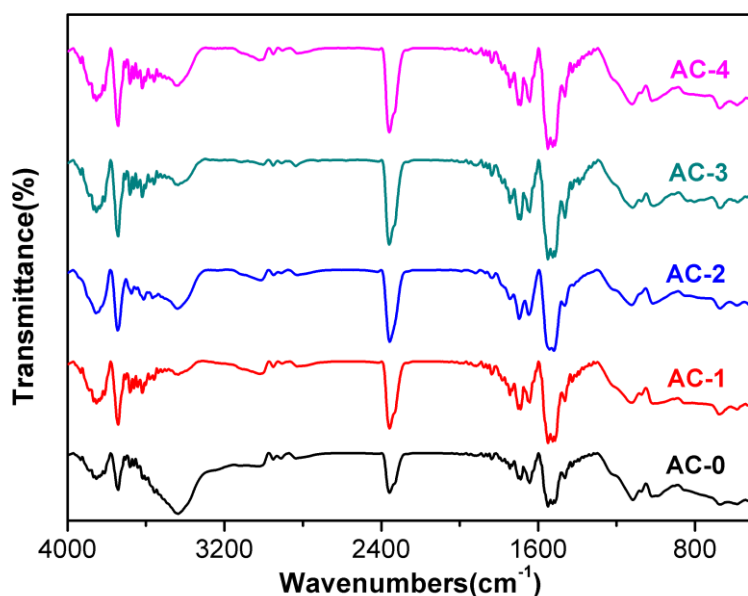


Fig. 4. FT-IR spectra of AC samples treated by SDAPPJs.

## 3.2. Chemical properties of AC

### 3.2.1. FT-IR analysis

Fig. 4 shows the FT-IR spectra of the AC samples. All spectra manifest a wide transmittance band at 3300-3600  $\text{cm}^{-1}$  and with a strongest peak at 3440  $\text{cm}^{-1}$ , it can be assigned to the O-H stretching mode of hydroxyl groups and absorbed water after sample modification[31]. And the sharp transmittance band at 3739  $\text{cm}^{-1}$  may attribute to isolated O-H groups[32].

The presence of absorption bands located at around 2700-3000  $\text{cm}^{-1}$  is due to O-H stretching vibration from carboxylic acid[33]. These two weak bands at 2841  $\text{cm}^{-1}$  and 2921  $\text{cm}^{-1}$  are caused by aliphatic group, i.e. C-H stretching in -CH-, and the characteristic deformation vibration peak of -CH- skeleton also can be found at 1450  $\text{cm}^{-1}$ [32]. The small band observed at about 1700  $\text{cm}^{-1}$  can be assigned to C=O stretching vibrations of carboxyl, esters or lactone groups[10, 34].

The FT-IR spectra also show a strong band at 1542  $\text{cm}^{-1}$  which is attributed to C=C vibrations in aromatic rings[32]. On its left, the olefinic C=C groups band at 1648  $\text{cm}^{-1}$  can be observed[35]. In

addition, the broad band at 1000-1300  $\text{cm}^{-1}$  is due to C-O stretching vibration from acids, alcohols, ethers or ester groups[36].

### 3.2.2. XPS analysis

XPS analysis was carried out to compare the detailed surface chemical composition between the untreated and SDAPPJs-modified AC for different time. The surface element content of AC and the percentage of various oxygen-containing functional groups are listed in Table 4. The oxygen content of SDAPPJs-treated samples is higher than pristine materials, with an increased O/C molar ratio from 7.93% to 12.39% after 3 min treatment. However, when the treatment time prolonged, the O/C ratio decreased to 7.88%. It was reported that the disordered fraction of carbon materials contains a huge number of defects that associate with high densities of unpaired electrons. Thereby it shows a strong ability to chemisorb other species, such as activated atoms or free radicals[39]. Therefore, it is quite easy to attach oxygen-containing active substances to the hanging bonds of AC, which is why the oxygen content is significantly increased after 3 min treatment. In addition, highly active species, high-energy electrons and thermal effect can promote the conversion of carbonyl groups to carboxyl groups[28].

Generally, adsorption performance can be improved in two aspects with the effect of functional groups. One is that oxygen-containing functional groups increase the surface polarity of AC, which would enhance the ability to adsorb polar molecules. The other is that the basal structural units of carbon surface can act as soft centers that might trap soft ions, whereas the oxygen-containing surface groups may become the hard sites that immobilize hard ions and molecules[37]. Therefore, the higher proportion of oxygen content, the stronger adsorption capacity of the sample.

Table 4. The molar ratio of carbon to oxygen and the C1s peak results of samples.

Samples	Element content (%)		O/C ratio (%)	Oxygen-containing groups			$\pi$ - $\pi$ transitions (%)
	C1s	O1s		C-O (%)	C=O (%)	O-C=O (%)	
AC-0	92.00	7.30	7.93	12.59	6.59	7.37	7.98
AC-1	88.00	10.90	12.39	16.46	5.76	9.42	8.93
AC-2	91.50	8.50	9.29	15.09	5.29	9.25	9.30
AC-3	92.60	7.30	7.88	15.24	5.50	10.08	8.42
AC-4	92.60	7.40	7.99	14.82	5.06	8.99	9.82

However, the extension of treatment time leads to the partial destruction of AC structure, which was verified by the N<sub>2</sub> adsorption isotherms and SEM images of AC. Regarding of the surface oxygen content, it also shows the trend of firstly increasing and then going down towards the opposite direction around the curve peak.

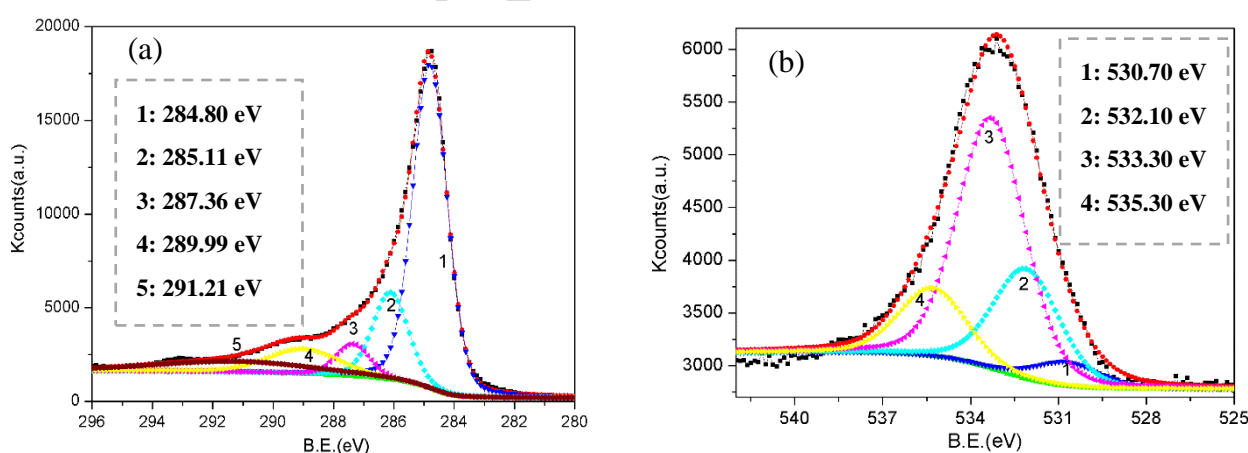


Fig. 5. C1s(a), O1s(b) deconvolution of the SDAPPJs-treated AC.

Fig. 5 depicts the C1s core level spectra of the SDAPPJs-treated sample (AC-1), the peak parameters and component assignments are given in Table 4. The C1s spectra of AC-1 has been fitted by five

components of the binding energies at approximately 284.80 eV, 286.11 eV, 287.36 eV, 289.99 eV and 291.21 eV, as marked in Fig. 5, namely (1) aromatic, graphitic or aliphatic carbon, (2) ether, phenol or alcoholic groups (C-O), (3) carbonyl groups (C=O), (4) carboxyl or ester groups (O-C=O) and (5) shake-up satellite peaks caused by  $\pi$ - $\pi$  transitions in aromatic rings. The results are well consistent with the previous studies[37-42]. On the other side, the O1s spectra at 530.7 eV, 532.1 eV, 533.3 eV and 535.3 eV correspond to (1) oxygen in carbonyl groups (C=O), (2) oxygen atoms in hydroxyl or ethers (C-OH or C-O-C), (3) oxygen in anhydride, lactone, or carboxylic acids (O-C=O), and (4) chemisorbed oxygen or water[12].

Through the peak fittings, it is found that the carbonyl groups do not changed dramatically after the treatment, however, the hydroxyl, ether and the carboxyl groups clearly increased. In addition, the more acidic functional groups on the surface of AC, the stronger its ability to adsorb MB molecules[10], which can be confirmed by the adsorption performance tests of AC.

### 3.3. Adsorption performance evaluation

AC shows a strong purification capacity to the sewage containing both organic polymers, such as dyes and phenolphthalein, and some inorganic substances including metal ions and acid, etc. In this work, MB was selected as a sewage candidate to test and verify adsorption performance of AC[43] and to evaluate the effect of SDAPPJs treatment on it. Usually, physical and chemical properties of AC surface play an essential role in the adsorption of both organic and inorganic substances.

MB is a kind of cationic dye and its dimensions are  $1.43 \text{ nm} \times 0.61 \text{ nm} \times 0.4 \text{ nm}$ [44]. Hence, it only can be adsorbed by micropores with large diameter, mesopores and macropores. Its adsorption by AC is very sensitive to the pH conditions of solution, when pH value is more than 7, the adsorption value



for MB increases with PH value. Generally, MB molecules are randomly vibrated in the aqueous solution to keep thermodynamic stability. Specifically, the whole adsorption processes can be described as follows: (1) MB molecules diffuse to AC surface from the solution; (2) MB molecules diffuse along the surface layers of AC and enter the inner channels along the transfer pores (macropores). When MB molecules encounter the active sites on the surface of AC, physical or chemical adsorption occurs; (3) MB molecules continue to move along the macropores and diffuse to the internal structure of AC, which finally are caught by active sites and appropriate micropores that conduct capillary adsorption with MB molecules. In general, the dominant step of the process is the diffusion of matters from the solution to the surface of AC. However, a recent study has reported that intraparticle diffusion step of MB molecules in AC was even slower[45].

There are two classical explanations with respect to the MB molecules adsorbed on AC. One is that cationic dyes can interact with the negatively charged functional groups on the AC surface due to electrostatic effect. The other is attributed to the dispersive interaction of MB molecules with the surface layers of AC, which was caused by the  $\pi$ - $\pi$  electron coupling occurred between the  $\pi$  electrons exist within the MB molecules and delocalized  $\pi$  electrons of AC[46]. Besides, to ensure that AC has a good adsorption capacity, enough specific surface area and suitable pore structure seem to be necessary. That is to say, with the increasing specific surface area and appropriate pore size distribution, the adsorption performance of AC can be improved eventually.

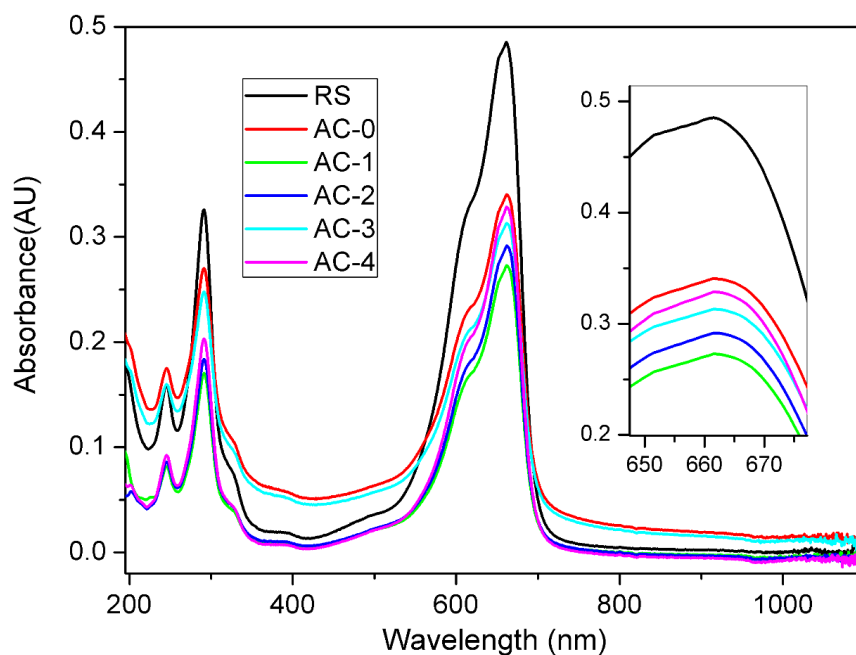


Fig. 6. UV-Vis absorption spectra of remained MB after adsorbed by samples.

Table 5. MB adsorption capacity of AC samples.

Samples	Absorbance (a.u.)	IC (mg/ml)	EC (ug/ml)	$q_e$ (mg/g)
RS	0.4786	0.0032	3.2000	None
AC-0	0.3406	1.6000	0.6832	458.41
AC-1	0.2691	1.6000	0.5398	530.11
AC-2	0.2917	1.6000	0.5852	507.41
AC-3	0.3131	1.6000	0.628	485.98
AC-4	0.3286	1.6000	0.6591	470.44

(IC: the initial concentration; EC, the end concentration;  $q_e$ , the equilibrium adsorption; RS, the reference sample)

In fact, the adsorption capacity of AC is affected by a combination of numerous factors. Comparisons of adsorption performance for AC samples prepared in this study are given in Fig. 6. The

only variant for these samples is SDAPPJs treatment time. It can be seen that the adsorption performance of the modified AC is better than pristine materials, and the adsorption capacity of AC-0, AC-1, AC-2, AC-3 and AC-4 are 458.41 mg/g (0.480 mg/m<sup>2</sup>), 530.11 mg/g (0.506 mg/m<sup>2</sup>), 507.41mg/g (0.501 mg/m<sup>2</sup>), 485.98 mg/g (0.495 mg/m<sup>2</sup>) and 470.44 mg/g (0.483 mg/m<sup>2</sup>), respectively. It is obvious that AC-1 possesses the best MB adsorption capacity compared to the other AC. Notably, the effect of modification is relatively more remarkable in the first 3 minutes, and the relevant adsorption performance of AC increases by 15.64%. However, when the modification time extends to 10 min, the MB adsorption capacity of AC only increases by 2.62%. In addition, the adsorption performance of AC treated by APPJSDs for 2 min and 4 min was also tested, and the amount of MB adsorbed were 522.79 mg/g and 515.37 mg/g, respectively. Both are less than the adsorption capacity of AC APPJSDs-treated for 3 min.

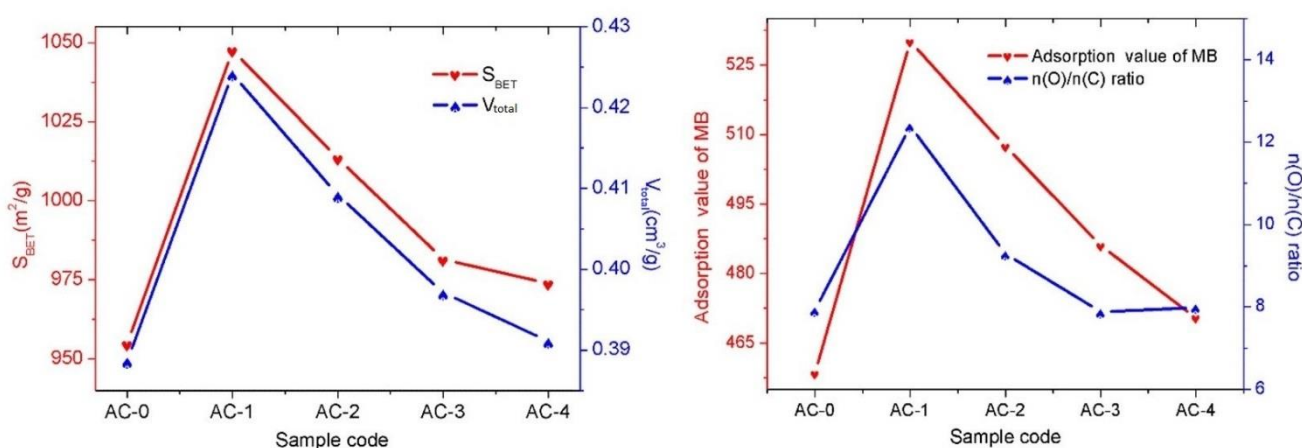


Fig. 7. The relationship between adsorption performance and properties of AC.

Furthermore, the specific surface area of AC comes up to the maximum value (1047.6 m<sup>2</sup>/g) through the first 3 min treatment, as same as the quantity of surface oxygen-containing groups. However, the oxygen content on the surface slightly decreases and the specific surface area reduces correspondingly

with the increase of the modification time, leading to the reduction of the amount of MB adsorbed (Fig. 7). Moreover, it is an interesting phenomenon that the amount of MB adsorbed increases from 0.480 mg/m<sup>2</sup> to 0.506 mg/m<sup>2</sup> through 3 min treatment. It is supposed that MB molecules can be randomly adsorbed on the active sites due to the electrostatic forces, in other words, such MB adsorption is primarily chemisorption[10, 47]. Furthermore, the increase of oxygen-containing groups on the carbon surface through SDAPPJs modification enhances electrostatic effect that would cause a molecular stacking effect, thus vertically locating MB molecules on the active sites[11].

### 3.4. SDAPPJs Modification Mechanism

APPJs can generate high concentration of active substances without vacuum at low temperatures as illustrated in Fig. 8.a. In such a system, porous AC is equivalent to the grounded electrode, therefore, when the distance between APPJs and AC is short enough, spark discharge occurs at the tip of the APPJs. Fig. 8.b indicates fresh pores can be generated on the surface of AC under the high-energy spark channels created by SDAPPJs, which also make the closed pores and dead end pores partly transformed into open pores or through ones. Moreover, the specific surface area of SDAPPJs-treated AC samples increases obviously but the pore size distribution changes little, which implies AC samples still possess good structural stability after SDAPPJs treatment.

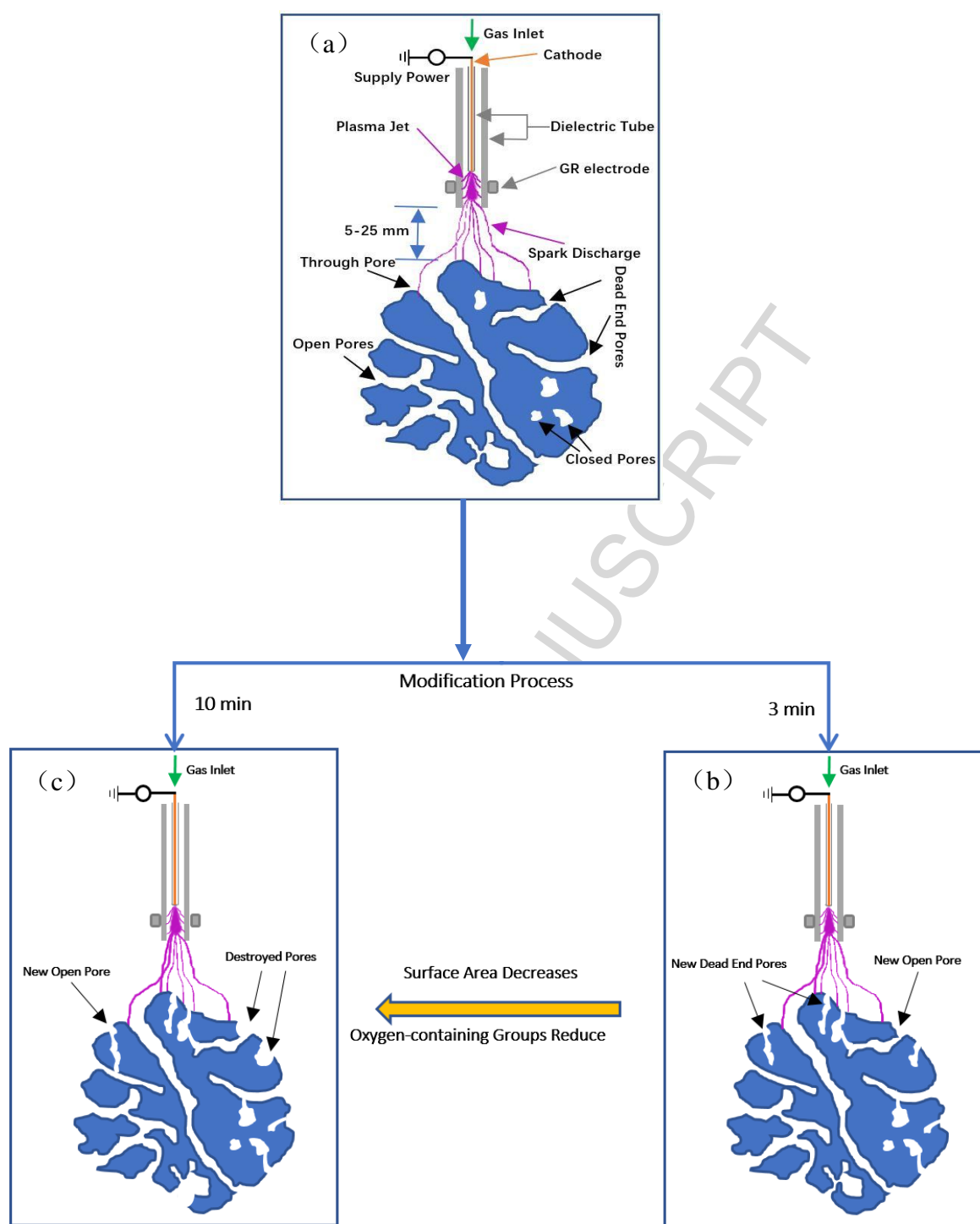


Fig. 8. Mechanism for SDAPPJs treatment of AC.

(a) equipment sketches of APPJs and the phenomenon of SDAPPJs when distance is about 5-25mm, (b) modification for 3 min, (c) treatment for 10 min.

In addition, these high-concentration active species produced by SDAPPJs could raise the number of oxygen-containing groups on AC surface that enhance the electrostatic interactions with cationic dyes. In this study, such active species are predominantly oxygen free radicals ( $\cdot\text{O}$ ), which would react with oxygen in the air immediately and then ozone produced. Thus, the formation of functional groups on the AC surface is mainly due to the role of ozone and the thermal effect caused by SDAPPJs. Specifically, it mainly results into the increase of hydroxyl groups as well as the conversion of carbonyl groups ( $\text{C}=\text{O}$ ) into carboxyl functional groups ( $\text{O}-\text{C}=\text{O}$ ). However, with further extended treatment time, specific surface area and oxygen-containing groups decreased. The possible reason for such phenomenon can be described in Fig. 8.c. The high-energy spark channels may devastate few pores on the surface of AC and part of the grafted oxygen-containing group bonds may be broken by the highly active species. Therefore, the MB adsorption performance of AC becomes relatively poorer compared to the one through 3 min SDAPPJs treatment.

#### 4. Conclusion

In summary, AC has been successfully modified by SDAPPJs treatment and characterized by  $\text{N}_2$  isotherms, SEM, FT-IR and XPS in this study. According to the analytical results, it is believed that SDAPPJs treatment can be a facile surface modification approach with advantages of low cost, high efficiency and less environmental impact, and it also gives a significantly positive effect on both surface physical and chemical properties of AC. SEM observations show that there are various in-situ pores generated on the AC surface after SDAPPJs treatment, which results in 9.71% and 9.13% improvement on the specific surface area and total pore volume, respectively. XPS results reveal that the O/C molar ratio on AC surface treated by SDAPPJs increases from its pristine state i.e. 7.93% to

12.39%. Specifically, the percentage of carboxyl or ester groups ( $\text{O}-\text{C}=\text{O}$ ) increases from 7.37% to 12.66%, and the percentage of ether, phenol or alcoholic groups ( $\text{C}-\text{O}$ ) raises from 12.59% to 16.46% after 3 min treatment. All these alterations to the physical and chemical properties cause a 15.64% improvement on MB adsorption capacity of AC in the sewage. The adsorption performance of modified AC is directly proportional to its specific surface area as well as the surface oxygen-containing groups. This work verifies the feasibility of SDAPPJs as a facile surface modification method of AC to obtain a significant enhancement on its adsorption performance, which brings attractive and fascinating application potentials in various practical aspects, such as sewage treatment and air purification.

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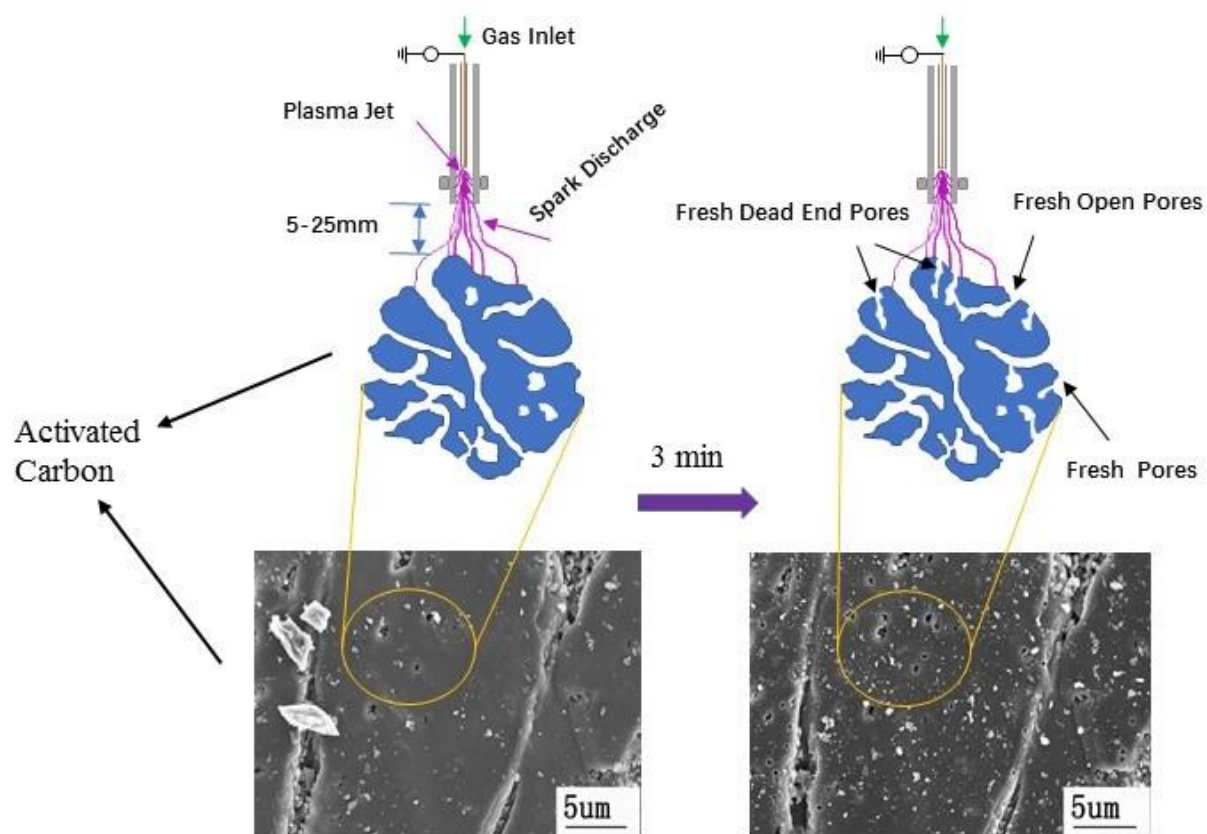
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Graphical abstract

## Highlights

- Activated carbon (AC) was modified by spark discharge of plasma jets.
- The textural and chemical properties of AC improved by the modification treatment.
- SEM observations show that various in-situ pores generated on the surface of AC.
- The treatment enhanced the adsorption capacity of AC to methylene blue.