

# Plasma–Solution Junction for the Formation of Carbon Material

Jiangqi Niu <sup>1</sup>, Chayanaphat Chokradjaroen <sup>1</sup>, Yasuyuki Sawada <sup>1</sup>, Xiaoyang Wang <sup>1,2</sup> and Nagahiro Saito <sup>1,2,3,4,\*</sup>

<sup>1</sup> Department of Chemical Systems Engineering, Graduate School of Engineering, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan

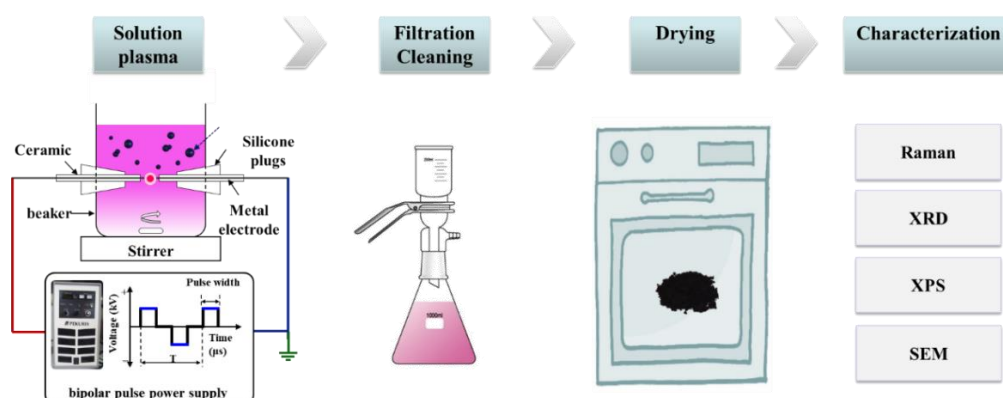
<sup>2</sup> Japan Science and Technology Corporation (JST), Strategic International Collaborative Research Program (SICORP), Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan

<sup>3</sup> Conjoint Research Laboratory in Nagoya University, Shinshu University, Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan

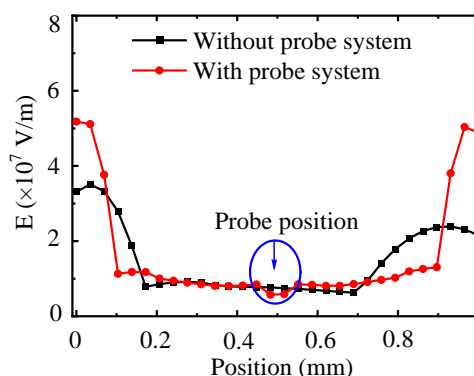
<sup>4</sup> Japan Science and Technology Corporation (JST), Open Innovation Platform with Enterprises, Research Institute and Academia (OPERA), Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan

\* Correspondence: hiro@sp.material.nagoya-u.ac.jp

The experiment operation process is shown in Figure S1. Firstly, we conducted the solution plasma discharge in the specific precursors under the bipolar power source. Then we collected the carbon by using vacuum filtration. Next step was dry the carbon products by using oven. Finally, the properties of carbon could be obtained by applying Raman, XRD, XPS and SEM.



**Figure S1.** schematic illustration of solution plasma process experiment, including solution plasma, filtration cleaning, separation drying and characterization of carbon products.



**Figure S2.** The electric field strength compared with or without Langmuir probe system.

The following three dimensionless parameters are key to determine the plasma sheath thickness and to apply the relevant computational model, including the electric Reynolds numbers ( $Re$ ), Debye ratio ( $\omega$ ), and a probe bias voltage ( $\chi$ ).

$$R_e = \frac{2r_p v_f}{\mu_i T_e} \quad (1)$$

$$\omega = \frac{\lambda_{DSP}}{v_p} \quad (2)$$

$$\chi = \frac{V_p}{T_e} \quad (3)$$

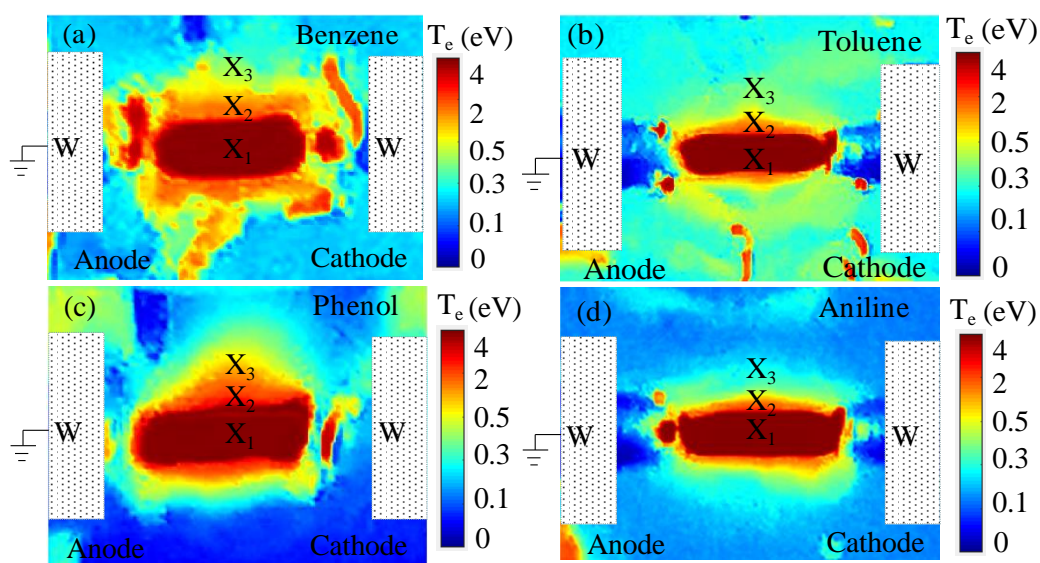
$v_f$  is bulk plasma flowing velocity,  $\mu$  is ions particles mobility,  $v_p$  is Langmuir probe DC bias voltage. It was found that the calculated criterion of the sheath convection mode in a wide range of operation conditions of solution plasma satisfied the following relationships:

$$R_e \omega^2 \chi^2 \gg 1, R_e \omega^2 < 1 \text{ and } \omega \chi \ll 1.$$

The ion current detected by the probe in this case comes from the coupled effect of the ion acceleration mechanism inside the sheath and direct convection. The plasma density ( $N_e$ ) and plasma electron temperature ( $T_e$ ) were calculated from the following equations according to the ion saturation current  $I_{isat}$  and the following formular:

$$n_e = \left[ \frac{I_{isat}}{5.3(e v_f v^{2/3} \mu^{1/3} \epsilon_0^{1/3} r_p^{1/3})^{3/4}} \right]^{4/3} \quad (5)$$

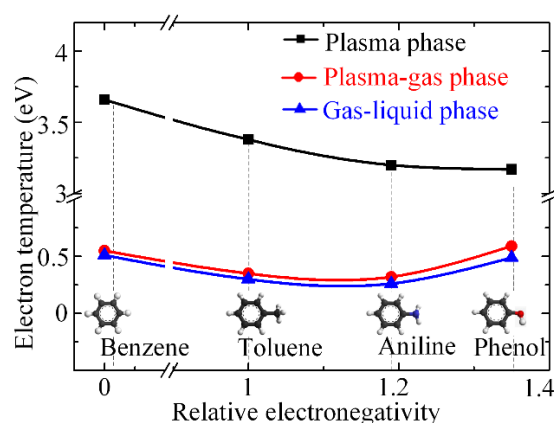
$$T_e = \frac{16 I_{isat}^2}{n_e^2 e \mu_i r_p v_f k_B} \quad (6)$$



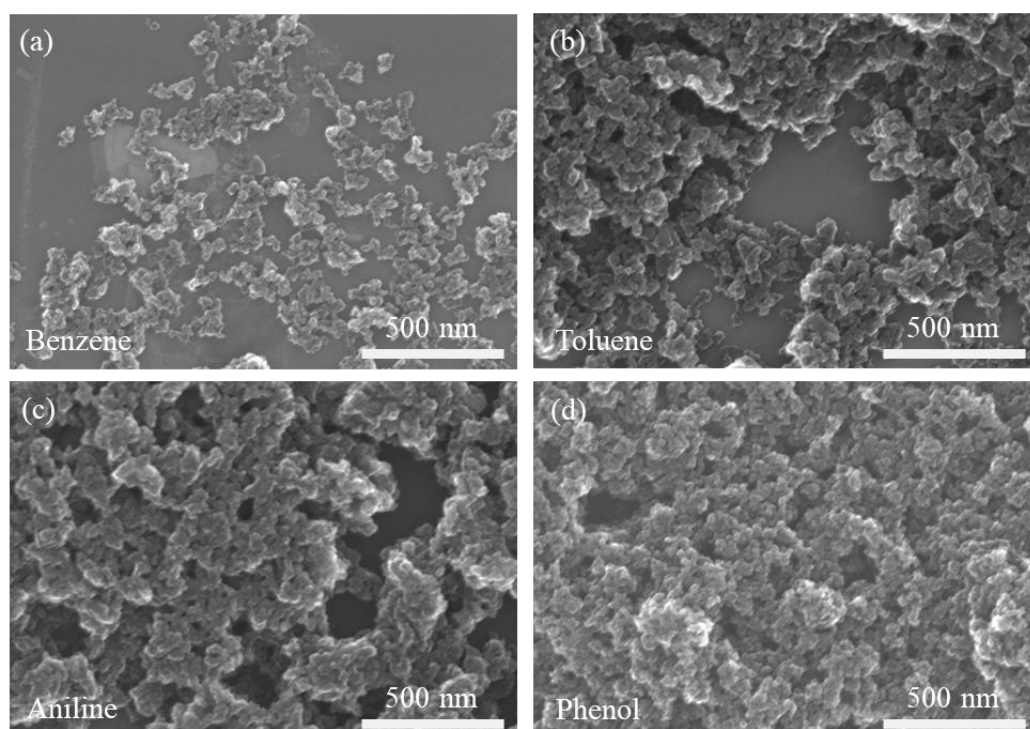
**Figure S3.** Temperature structure characteristics of the negative polar discharge plasma in four solutions of (a) benzene, (b) toluene, (c) phenol, (d) aniline mapping.

**Table S1** Bulk and surface elemental composition of products from benzene, toluene, phenol and aniline obtained from XPS measurement; Nitrogen bonding composition of product from aniline.

Precursor	XPS (at%)			Nitrogen bonding composition (%)		
	C	N	O	N <sub>pyridinic</sub>	N <sub>pyrrolic</sub>	N <sub>quaternary</sub>
Benzene	92.12	-	7.88	-	-	-
Toluene	84.1	-	15.89	-	-	-
Phenol	80.95	-	19.05	-	-	-
Aniline	88.78	3.53	7.69	9.94	73.73	16.32



**Figure S4.** Variation of electron temperature with relative electronegativity (C=2.55, N=3.04, O=3.44 / C=2.55)) at different interfaces of solution plasma.



**Figure S5.** SEM images carbon products of (a) benzene, (b) toluene, (c) aniline, (d) phenol.