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Electrical discharge of butane and propane for carbon plasma with air impurities, slightly ionized at high pressure

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ABSTRACT

This paper investigates the ionization of butane and propane gas in the formation of carbon plasma with air impurities. The formation of carbon plasma is designed and operated by using a commercial pencil graphite electrode with a directionally electrical discharge method of less than 3.5 kV, injecting butane and propane gas with a mixture of air, pressure variations into the tube. Experimental results show the voltage and current before and after the breakdown voltage. The density and temperature of the plasma carbon are calculated from the current and the breakdown voltage. These results depicts that the ionization of air at a pressure of 12 cmHg (570 V) produces a breakdown voltage of 530 – 570 V and current of 0.27 – 0.45 mA, whereas at pressure of 37 cmHg (1160 V) produces breakdown voltage of 900 – 1100 V and current of 0.46 – 0.6 mA. The ionization of the butane and propane (with impurities of air) at pressure of 26 cmHg (1199 V and 1137 V) produces breakdown voltage of 831 – 1202 V, 839 – 1138 V and current of 0.07 – 0.65 mA, 0.08 – 0.63 mA. At the pressure of 46 cmHg (1828 V and 1590 V) produces breakdown voltage of 1246 – 1781 V, 1021 – 1627 V and current of 0.04 – 0.99 mA, 0.05 – 1.12 mA. These results indicate that the carbon plasma density increases with the increase of electrical voltage. This report shows good agreements for developing the growth of carbon nano tube on the surface of cathode which is generally always followed by air impurities at high pressure.

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1. INTRODUCTION

The development of atmospheric plasma and technology research is nowadays increasingly important in industries such as steel cutting, welding, as a source of energy and the development of materials, especially to grow carbon nano tubes (CNTs) in the cathode surface as well [1, 2]. Particularly in the growth of CNTs, the effort to ionize the gas, produce and grow CNT remains a research choice today. Many research results are found, such as a mixture of helium-argon gases has been carried out to generate plasma by arc-discharge technique at atmospheric pressure using a mixture of nickel and yttrium catalysts. This ionization process produces single-walled-nanotubes (SWNTs) purified by oxidation and centrifugation techniques [3-5]. CNTs have attracted great attention due to the demands of nanotechnology, conservation and storage energies, and large nanosensors. CNTs have been produced effectively by arc-discharge methods at atmospheric pressure using a gas mixture and are essential for producing high density carbon species [6-8]. The degree of purity of ionization is very important to be observed in forming CNTs, especially ionization products with high purity carbon species in generating CNT density in their structural and geometric diversity. The gases used are methane, argon, helium and argon + helium [9, 10] mixtures. Another example is the method of arc discharge method, laser ablation, HIPCo, molten salt technique and chemical vapor deposition (CVD) [11-13]. The arc discharge method is the first method used in finding CNTs and this method is the easiest and most efficient method of generating carbon ion ionization [14-17].

Given the impurity of ionized gases causing source and media factors greatly affect CNT formation. The source of the voltage breakdowns on ionization will experience some contribution and resistance to the formation of carbon ions. At a small electrode distance and diffusion length will also deliver ionizing effects that require varying energy. The DC voltage source is expected to deliver a positive carbon ion to the negative cathode so that its mobilization can be formed in large quantities and regular structures and geometries. Under the same conditions high voltage power and the impurity of air in the alkane group gas will increase the higher the breakdown voltage. In the case of a high-density gas, the collisions of gases mostly produce elastic collisions or electron excitations in atoms amongst particles, and polarization of dielectric gases by an electric field. This direct current voltage source (DC) will inhibit ionization while the movement of the gas particles will also be hampered not only because of the gas crossover, but also the atomic and molecular polarization effect reduces the kinetic velocity of the particles so that the kinetic energy and potential energy for ionization will decrease.

Therefore, although experimentally the phenomenon of breakdown voltage has been successfully understand because it works at the state of before and after the ionization, but in the understanding of the processes and effects of the particle gases diversity, at high pressure and DC voltage remain the research concern for an increase in the number of CNTs formed. In this experiment, a small plasma reactor tube contains an atmospheric pressure carbon gas with two electrodes connected to a voltage source. The carbon gas will be ionized and produce plasma discharge then the density and temperature can be calculated. This density and temperature analysis can be utilized to predict the growth of CNTs, their purity, geometry, structure and chain formation.

2. MATERIALS AND METHOD

2.1. Ionization of Carbon Plasma on Butane, Propane and Air Mixture

Ionization of gas by electron impact collision is very significant requirement to produce plasma at high pressure [18]. Kinetically, plasma can occur when the temperature or energy of the gas is increased, followed by the electrons out of the orbit. The magnitude of the ionization energy of an atom or molecule can be written in Equation (1):

$$W_E + \frac{1}{2}mv^2 + E_p \geq eU_i \tag{1}$$

where, m is the particle mass, v is the velocity of the particle, W_E is the effort due to the electric field, E_p is the electrical potential energy, e is the charge of the electron, and U_i is the ionization potential.

The process of plasma is influenced by several factors including the kinetic energy of atomic, molecular and photon collisions; Thermal and acoustic mechanical energy from external sources also can accelerate atomic/molecular motion to excite electrons until ionization. Another factor is the potential energy source contributed by the electric field and the external magnetic field thus accelerating the ionization process. The ionization process is also accelerated by involving both kinetic energy and potential energy. The ionization process, arc discharge can occur with two electrodes gap and high electric potential difference through the medium gas that can described by collision as follows $(N_2+O_2+CO_2+\text{trace})_{air}$ + C_4H_{10}/C_3H_8 → ionization (Air + Butane/Propane). The high voltage causes the polarization current and the kinetic collision leads the excited electrons to ionize to form ions [19]. Although the ambipolar diffusion process of polarization inhibits kinetic motion, high electrode voltage difference, electrode distance and short diffusion length will assist the ionization process to occur along the degree of ionization of butane or propane which has low ionization energy. Butane has a large density, large cross section, relatively small velocity and ionization energy of 10.6 eV which contribute ionization. The ionization energy required for each molecule in the butane/propane gas for ionization is a vector quantity that can be defined as follows:

$$\begin{bmatrix} f(E_1) \\ f(E_2) \\ f(E_3) \\ \vdots \\ \vdots \\ f(E_n) \end{bmatrix} \begin{bmatrix} E_{p1} & E_{p2} & E_{p3} & \dots & \dots & E_{pn} \end{bmatrix} = \begin{bmatrix} E_i(N_2) \\ E_i(O_2) \\ E_i(CO_2) \\ \vdots \\ \vdots \\ E_i(gas_i) \end{bmatrix} \tag{2}$$

The energy distribution function of each atom or molecule will collide for each particle and contribute diverse ionization energy. Therefore, by knowing the distribution of this energy, the ionization potential will easily form the plasma until the breakdown voltage occurs

2.2. Electrical Discharge with Direct Voltage, Thermal Plasma

The ionization process is carried out by increasing the voltage gradually so as to form plasma with voltage and electric current. Figure 1 is an electrical discharge schematic of the gas ionization tube. A DC voltage source is provided by connecting a carbon electrode (commercial pencil) about order of 1mm scale. Voltage is launched until the electrical spark is reached and maintained the arc discharge condition.

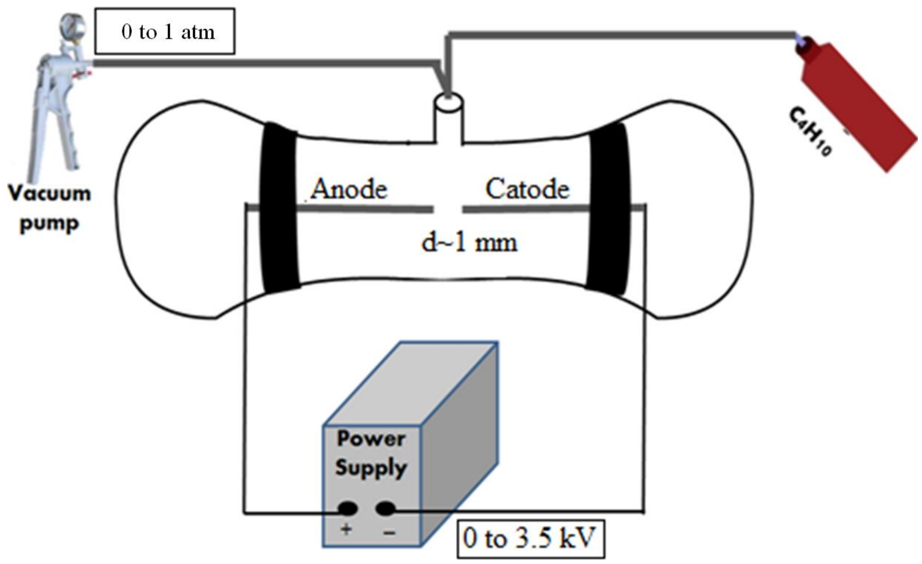

Figure 1. Design and operation of directional carbon plasma.

The air in the cylinder is reduced in the order of 0.1 atm which then occurs the breakdown voltage of any volume reduction in air pressure. The electrical current is maintained only at the order of 10 mA in order to keep the arc discharge conditions. The butane and propane gases are also alternately streamed until 1 atm is subtracted to obtain the breakdown voltage phenomenon until the air density becomes smaller as a form of impurity of ionized air, dominated by alkane gas. Butane and propane are used interchangeably and not mixed in the same process. The butane gas or propane which is flowed with the air gas is in a random velocity. This speed can be known statistically by using the Maxwell-Boltzmann equation:

$$f(v) = \frac{dn_v}{dv} = \frac{4n}{\sqrt{\pi}}\left(\frac{m}{2k_BT}\right)^{\frac{3}{2}} v^2 e^{\left(-\frac{mv^2}{2k_BT}\right)} \tag{3}$$

The velocity distribution of gas particles in the tube can be shown in Figure 2. Physically, the kinetic and thermodynamic contributions strongly dominate the event of the breakdown voltage with the electric field effect that atomic and molecular polarization speeds up the ionizing process.

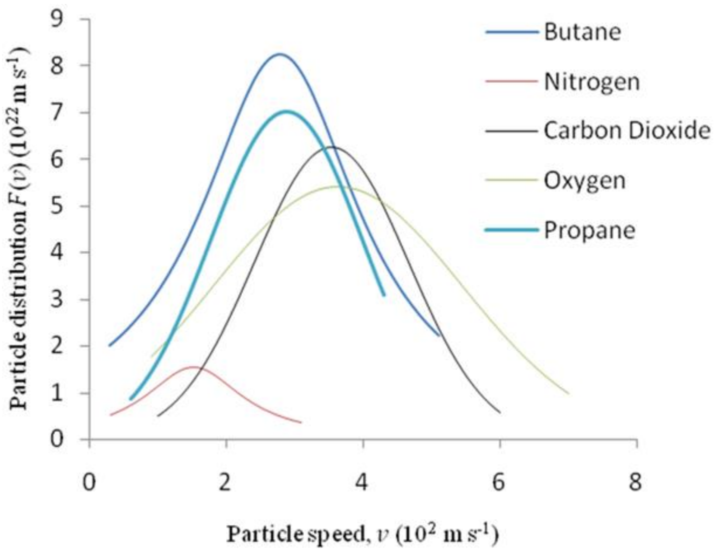

Figure 2. Gas particle velocity distribution at atmospheric pressure.

Plasma density and temperature of carbon species with general air impurities can be expressed as follows:

$$n = \frac{2\varepsilon_0 \varepsilon_r V}{ed^2} \quad (4)$$

$$p = nk_B T \quad (5)$$

In the event of this thermal plasma, not all particles are fully ionized so that the density and temperature depend upon the source of the electrical voltage, the effect of the electric field between the electrodes, the high particle velocity and the mean free path of particles is short, the particles are facilitated readily to be recombination and association, although this is highly desirable condition in the formation of CNTs on the cathode surface. Therefore the voltage drop varies greatly with the gas pressure of the alkane by the impurity of the air mixture.

3. RESULTS AND DISCUSSION

Ionization of air in the cylindrical tube mainly imposed by electric field is successfully carried out by varying high pressure. At 12 cmHg (570 V), it produces the breakdown voltage of 0.53 – 0.57 kV and current of 0.27 – 0.45 mA. The pressure of 16 cmHg (580 V) produces the breakdown voltage of 0.49 – 0.56 kV and the current of 0.31 – 0.4 mA. The pressure of 26 cmHg (920 V) produces the breakdown voltage of 0.86 – 0.88 kV and the current of 0.54 – 0.6 mA. The pressure of 36 cmHg (1160 V) produces the breakdown voltage of 0.9 – 1.1 kV and 0.46 – 0.6 mA. This data shows a nearly linear trend between the pressure with the current and breakdown voltage. The DC voltage given is relatively high because the gas ionization is at high pressure and the density of the particles inside the tube is also higher than 10^{24} particles/m^3. Electrical current from electron particles and positive ions moves successively to the anode and cathode. Movement of electrons to the anode is assumed to be faster than that of the cathode because the electrons are lighter with high density and speed, while the carbon positive ions move slowly toward the cathode (to form CNT) where the ambipolar diffusion is ignored. This movement is due to the influence of the potential difference gap between electrodes. When the gas pressure inside the tube is reduced then there is also a reduction in the density [20]. The particle density also affects the resulting breakdown voltage as shown in Figure 3. High densities result in collisions occurring predominantly at the excitation level of atoms and electrons so that ionization is difficult at high pressure because some of the energy is absorbed for elastic collision even superelastic collisions of each particle [21].

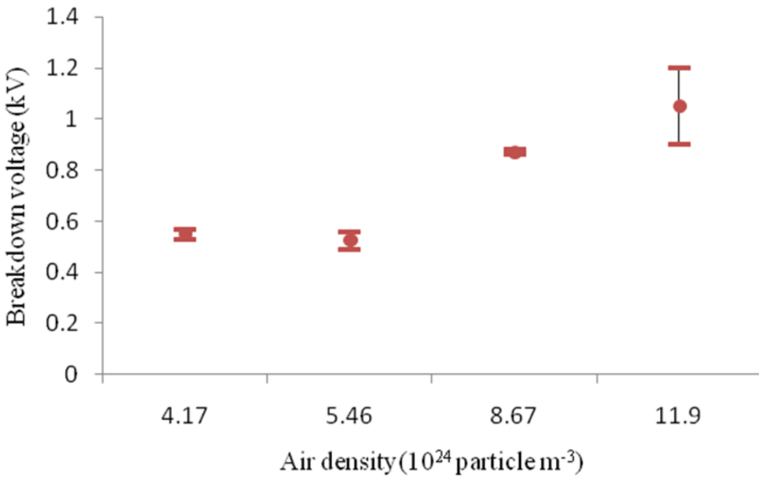

Figure 3. Air ionization with variation of air pressure.

Figure 4 shows a nearly linear kinetic relationship, although not all of all particles are ionized, this kinetic quantity is in thermodynamic equilibrium. Increased plasma density is followed by increased pressure so that plasma temperature will also increase. This temperature is the movement of electrons and ions in which both particles remain at the same temperature. At a pressure of 13 cmHg, the density of electrons and ions is relatively low, the pressure is small so that the collisions are small and the temperature decreases [22].

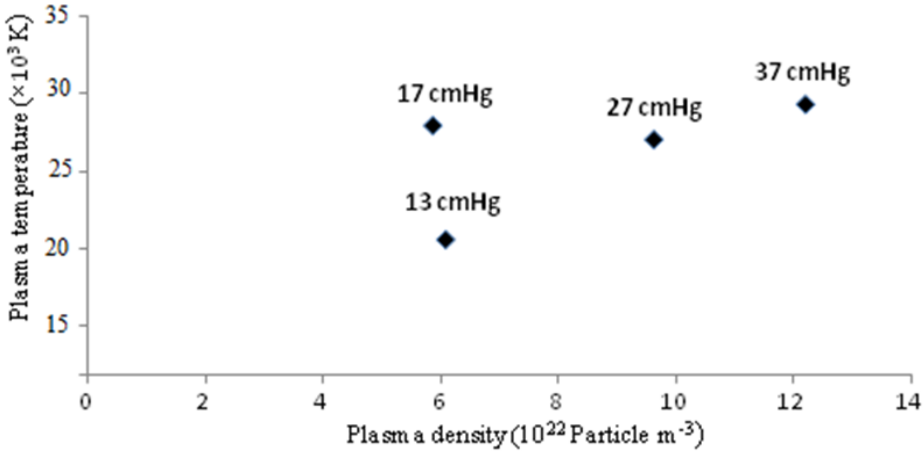

Figure 4. Air ionization for some pressure.

The ionization of the (butane, propane) gas is respectively subjected to the same variation of pressure to depict the resulting ionization difference. At a pressure of 26 cmHg (1199 V and 1137 V) it has a particle density of 8.29 × 10^{24} particles/m^3, the breakdown voltage of (831 – 1202 V and 839 – 1133 V) and the current of (0.07 – 0.65 mA and 0.08 – 0.63 mA). At a pressure of 31 cmHg (1303 V and 1165 V) it has the particle density of 9.98 × 10^{24} particles/m^3 producing the breakdown voltage of (950 – 1303 V and 827 – 1179 V) and the current of (0.07 – 0.71 mA and 0.03 – 0.46 mA). The pressure of 36 cmHg (1465 V and 1335 V) has the particle density of 11.5 × 10^{25} particles/m^3, resulting in the breakdwon voltage of (1064 – 1509 V and 938 – 1341 V) and the current of (0.04 – 0.75 mA and 0.02 – 0.77 mA). The pressure 41 cmHg (1648 V and 1516 V) has the particle density of 13.1 × 10^{25} particles/m^3 resulting in the breakdown voltage of (1180 – 1717 V and 1007 – 1507 V) and the current of (0.04 – 0.98 mA and 0.04 – 0.84 mA). The pressure of 46 cmHg (1828 V and 1590 V) has the particle density of 14.9 × 10^{25}particles/m^3 resulting in the breakdown voltage of (1246 – 1781 V and 1021 – 1627 V) and the current of (0.04 – 0.99 mA and 0.05 – 1.12 mA).

The applied voltage for ionization of propane gas and butane gas with air mixture tends to be greater than the total air gas due to increased butane and propane density, relatively slow particle velocity, and the area of the gas particle cross section tends to be large. These results indicate to ionize propane gas has a small voltage compared to butane because propane has a density and a smaller cross-sectional area than butane.

Figure 5, 6, and 7 describe the resulting breakdown voltage to the change in the density of the particle by comparing the ionization occurring between the mixture of propane + air and butane + air.

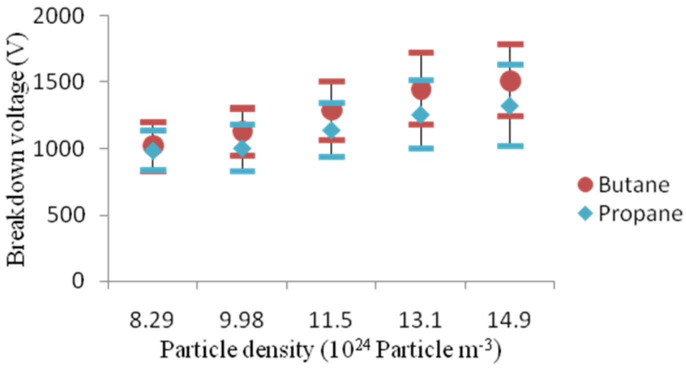

Figure 5. Ionization of propane and butane gas with air mixture.

Figure 5 shows the ratio of breakdown voltage generated by the butane and propane gas. Propane breakdown voltage is not only contributed to the high electric field, but also the greater carbonized ionization opportunities. Propane has ionization energy of 11.1 eV and butane 10.6 eV. The ionization of propane gas requires a small voltage rather than butane even though the ionization energy is large. The propane density is small compared to butane as the propane particle velocity is relatively larger than the butane. From Equation (1), the difference between the two ionization alkanes lies in the kinetic energy, the speed and the thermal propane are greater than the butane has [23].

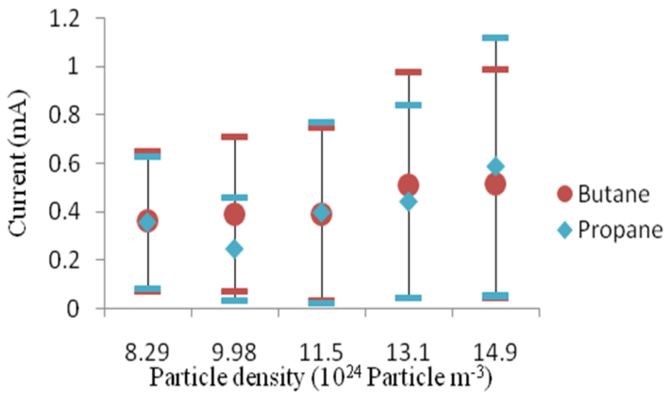

Figure 6. Discharge of propane and butane gas with air mixture.

Figure 6 describes the electrical discharge process between the two electrodes. Electrons move at speeds that are affected by electric potential, electric fields, drifting collisions, and collision frequencies that occur during the ionization process. The energy given to ionization is made up of mechanical energy and electromagnetic energy. The electric current is maintained from the effects of the system in order to maintain the balance of the arc discharge by particle collisions for not being recombination or neutral particles so that the plasma conditions remain to exist in a few seconds [24].

The greater the plasma density is calculated in the tube, the higher the plasma temperature is reached as depicted in Figure 7. This is also followed by the addition of the gas pressure. The result is smaller than the ionization in air due to the relative butane mass greater than the air particles while the relative masses of propane are smaller than air. The carbon chains of butane and propane gas are also large so that ionization obtains easier. Ionization of butane has a low temperature compared to propane, due to the effect of un-ionized hot gas making it easy to recombine the particles to be neutral, the greater butane cross-sectional effect and high propane dissociation effect so that the propane temperature is higher [25].

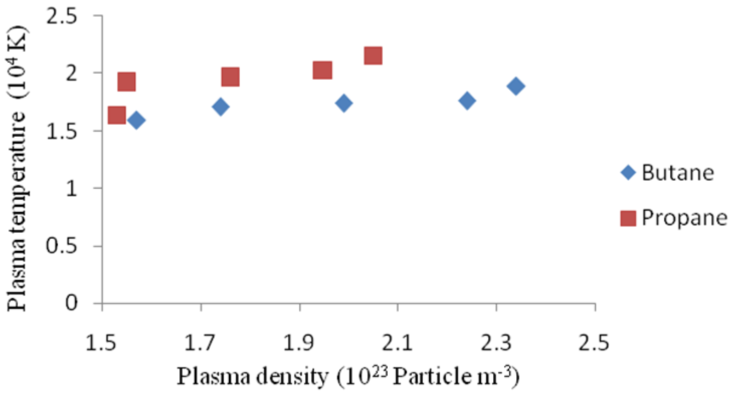

Figure 7. Density of alkane particles in thermal plasma.

4. CONCLUSION

The ionization of the Carbon produced by the butane and propane gas has a linear trend. The design and operation of carbon plasma has successfully conducted. Respectively, the ionization of (butane, propane) gas at a pressure of 26 cmHg (1199 V and 1137 V) has a particle density of 8.29×10^{24} particle/m^3, the breakdown voltage of (831 – 1202 V and 839 – 1133 V), a current of (0.07 – 0.65 mA and 0.08 – 0.63 mA), density 1.57×10^{23} particle/m^3, 1.57×10^{23} particle/m^3 and temperature (1.59×10^4K and 1.64×10^4K). Pressure 46 cmHg (1828 V and 1590 V) has a particle density of 14.9×10^{25} particles /m^3, resulting in the breakdown voltage of (1246 – 1781 V and 1021 – 1627 V), the current (0.04 – 0.99 mA and 0.05 – 1.12 mA), density is (2.34×10^{23} particle/m^3 and 2.05×10^{23} particle/m^3) and temperature (1.89×10^4K and 2.16×10^4K). This plasma density and temperature of butane and propane have been calculated. The ionization has an additional voltage when the gas density increases. The butane and air mixture requires a great pressure of propane and air mixture for ionization because of the cross-section, ionization energy, and relative mass of the gas. The propane air mixture has a large plasma temperature compared to the mixed air butane ionization. The plasma ionization in this research is a reactive alkane gas with impurities of air mixture so that the resulting CNT product is not pure to the carbon composition.

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