FABRICATION AND CHARACTERIZATION OF CARBON COMPOSITE FROM COCONUT SHELL CARBON

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ABSTRACT

Structure and chemical composition of coconut shell carbon with polyvinyl alcohol (PVA) as the stimulant through the observation of TG-DTA, SEM-EDS and FTIR has been studied. The process was carried out by calcining coconut shell charcoal at the temperature of 873 and 1023 K under Nitrogen flow, then mixed with polyvinyl alcohol (PVA) under composition of 2.5 to 7.5% (wt) in water solvent. The growing of carbon composite structure was observed by heating the samples in Argon gas of 1673 K, the rate of temperature was 10 K/min in 3 h. The products were then analyzed by TG-DTA, SEM-EDS, FTIR and XRD. The result showed that the products were in uniform particle sizes of micrometer dimensions and spherical particles shape, with average content of C element was 97.44% (wt), aromatic character and semi-crystalline structure.

Keywords: *coconut shell carbon, PVA, carbon composite, TG/DTA, SEM/EDS, FTIR, XRD*

INTRODUCTION

Carbon materials can be used in various applications, such as electrodes, catalyst supports, adsorbents and filters. Carbon material (including composite materials containing carbon) are well-known for their structural applications, which relate to aerospace structures, aircraft brakes, concrete structures and lubrication [1]. The advanced carbon materials such as synthetic carbon composites obtained from carbon fibers struggle to be accepted in wider practical use. Of well known carbon materials, the role of composite carbon cannot be overestimated either in modern industrial practice or in everyday life. It happens due to the unique physical and chemical characteristics of composite carbon which attract researchers to conduct certain research which explores those characteristics [2].

The structures of chemical bonds that are formed contribute the establishment of superior properties of carbon materials [2]. Carbon contains allotrope which different types of chemical bonding structure, including graphite, diamond, black carbon, fullerene, carbon nano tubes (CNT). Graphite is a type of carbon material obtained from carbon atoms with sp^2 orbital. One of these atoms forms a new bond with 3 carbon atoms. The microscopic material consists of graphite flat sheets from carbon atoms which are bonded, as the graphene. The characteristics of the bond structure and dynamic interactions between the graphene layers produce strong electrical conductivity properties [4] which functions as a lubricant.

The heating process by using various methods has conducted to accelerate the adsorption of electron [4]. The mechanism can be as the simple as removal of surface impurities, however it may also involve the removal of oxide and even the structural rearrangement [5]. The using temperature from 1000 °C to 1720 °C results in the increasing of ratio of graphite layer in the crystallites from 4.5 to 33 and their diameter growing bigger from 1.8 to 6.3 nm.

Carbon used in the industry is an example of carbon that can be heated at temperatures up to 1000–1300 °C, while graphite is a type of carbon which can be heated at temperatures higher than 2500 °C. Heating at higher temperature of 3000 °C will produce carbon that has an irregular structure and it evolves more ordered graphite structure by removing the impurities of volatile material [6].

Composite carbon can be obtained by carbonizing coconut shell, and polyvinyl alcohol (PVA) in a thermal decomposition process [7-8]. The main composition of coconut shell consists of cellulose, lignite, and hemicelluloses with a formula C, O, H, and N. These organic materials contain functional groups such as hydroxyl (R-OH), alkanes (R-(CH_2) nR’), carboxyl (R-COOH), carbonyl (R-CO-R’), ester (R-CO-OR’), linear and cyclic clusters ether (ROR’) with a variation of carbon [9]. In this process, the cellular anatomic features of coconut shell and polyvinyl

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alcohol are retained in the new carbon material [2]. The structural changes during carbonization and the physical properties of the resulting chars have been investigated in several studies.

Polyvinyl alcohol (PVA) as an inert and organic adhesive is used to obtain high density products. PVA was chosen due to its homogeneity in functional groups, linearity and solubility in water [10]. With increasingly high-temperature treatment of adhesive polyvinyl alcohol (PVA), it further affects the growth of grain to be larger in size.

This study used coconut shell charcoal as raw material which functions as carbon to examine the growth of crystal in the development carbon composite. To obtain carbon from coconut shell charcoal, tar impurities and volatile material are removed. The removal of impurities is conducted by high temperature heating up to 600–1000 °C. This heating is also aimed to eliminate the compounds carried by the drainage of gas. During the heating process, all non-carbon materials must be removed to form a pure carbon as well as to arrange the structure [11-12]. Whereas, the removal of metals can be carried out by soaking them in to a solution of HCl [13].

Heating process of coconut shell will produce a gradual change. In the first stage of the carbonization, the carbon samples become coke. The second stage is the stage of graphitization in which it changes coke into graphite carbon whose irregular structures tend to grow over the graphite whose structure is irregular [14-15]. The nature of carbon is depends on the source of carbon [16], in addition to the method and conditions of synthesis [17]. The nature of carbon materials gives impact on their various uses, primarily as carbon electrodes and carbon material structure.

The recent study which particularly discusses on these materials, coconut shell and polyvinyl alcohol (PVA) are used to produce carbon composite materials (carbon bio-composite). The main purpose of this research is to study the structure and chemical composition by using parameters such as concentration of polyvinyl alcohol and sintering temperature on the crystal structure growth of carbon composite.

EXPERIMENTAL SECTION

Materials

This study used charcoals as the raw material produced by PT Tropica Nucifera Industry Bantul Yogyakarta, Indonesia. This charcoal is the source of carbon and polyvinyl alcohol. Further, it uses Merck stimulant and argon gas used as inert atmosphere to get air free.

Instrumentation

Calcination uses a tube-Thermolyne Furnaces (Sybron) Type 21100 with a maximum temperature 1200 °C. Calcinated charcoal as raw material was obtained through 100 mesh grinding and sifting. For carbon materials, it was used Tarno compacted Grocki 312 model a maximum of 20 tons. Sintering was carried out by using a Carbolite furnace-Edwards Pirani 501 A6D 1600 °C maximum temperature with argon gas as the atmosphere, Scanning Electron Microscopy (SEM) JOEL JSM-6360 LA and Energy Dispersive Spectroscopy (EDS) system JOEL JED-2300, and thermal reaction with TG/DTA METTLER. To study the spectrum of carbon materials, it was used Fourier Transform Infra Red (FTIR) Shimadzu model IR-Prestige 21 and X-ray Diffraction (XRD) analysis using Goniometer model diffractometer with Cu K_α (α = 1.54056 Å) radiation.

Procedure

Preparation and Charcoal Calcination

Carbon powder 100 mesh sieves was calcined in an inert state through two stages: calcination at temperatures of 873 K for 3 h with the existence of nitrogen gas then, it is followed by the purification charcoal powder which was extracted for 24 h with 1 M hydrochloric acid at room temperature [14]. The sample was washed with distilled water until it reached constant pH and was dried in an oven at temperatures at 383 K overnight [13]. Re-calcination was carried out for 3 h at temperature of 823 K with nitrogen.

The process of carbonization

The carbon powder was calcined with 2.5 to 7.5% (wt) polyvinyl alcohol then suspended in distilled water at 353 K and stirred for 60 min. Binder system was mixed with charcoal powder, forming a paste with the density in different systems. The next steps molding by using a cylindrical mold with a diameter of ~ 15 mm. Compaction was performed with one direction force by the means of Tarno Grocki with the thrust of 5 tons. In this stage, the process produced a sample of pellets (green compact). The samples were then dried at room temperature for one day before performing the drying process in oven for 4 h at 383 K. The samples were then fed into a carbollite furnace for sintering at the temperature of 1273 K. The rate of temperature was 10 K/min and reaction time was 3 h. Then, these samples were cooled in the furnace at a rate of 8 K/min, with a flow of argon gas. This process produced dense coconut shell coke or carbon-carbon composite materials [11,15,18].

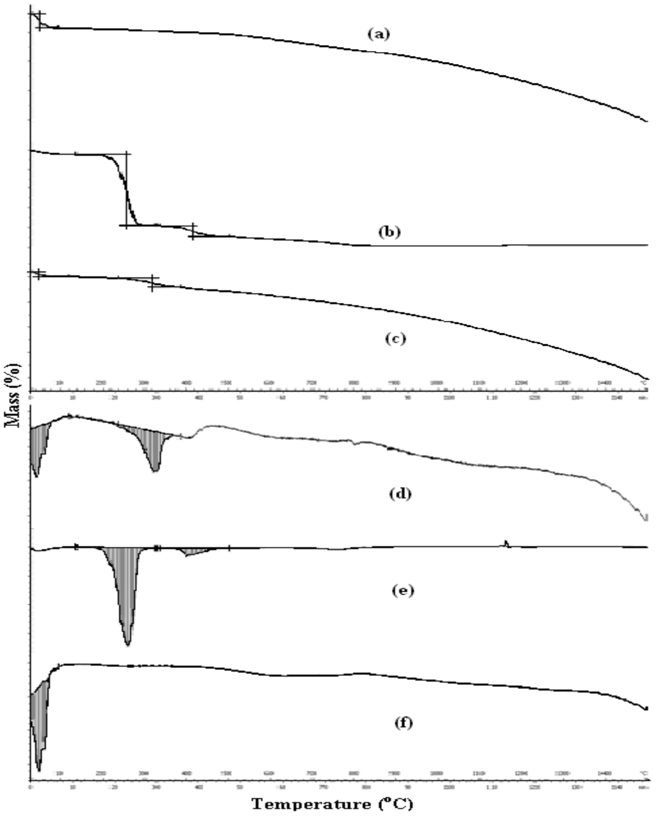

Fig 1. TG-DTA thermogram : (a,e) PVA a pure, (b,d) coconut shell carbon/PVA 7.5% (wt), (c,d) coconut shell carbon

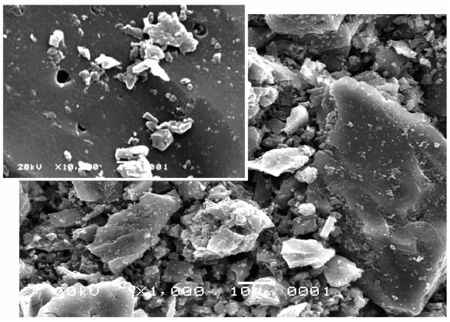

Fig 2. SEM micrographs of coconut shell carbon calcined at 1023 K

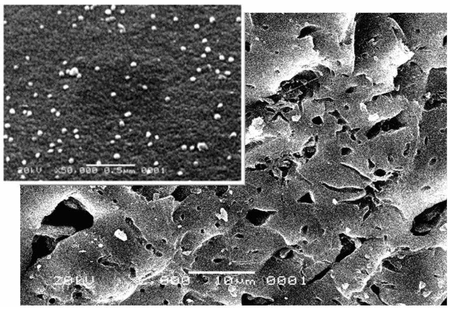

Fig 3. SEM micrographs of coconut shell Carbon/PVA (5% (wt)) sintered at 1673 K

Synthesis of carbon structures

The coke sample was heated for the 2nd time by sintering at 1473 K, 1673 K with a flow rate of 10 K/min, holding time of 3 h, in the furnace Carbollite, with the furnace cooling rate of 8 K/min, under argon gas flow [18].

RESULT AND DISCUSSION

Thermal Analysis

Fig. 1 showed TG/DTA measurements for the coconut shell carbon, polyvinyl alcohol and carbon-PVA mixed with various compositions at temperatures ranging from 30 to 1500 °C. Fig. 1(a) showed DTA curve of the coconut shell carbon with endothermic reaction at peak temperatures of 48.20 °C when the water is dissolved. The pure polyvinyl alcohol was obtained by the two endothermic peaks shown in Fig. 1(b). The decomposition of material had occurred in polyvinyl alcohol under phase I of water molecules evaporation at the peak temperature of 255.76 °C and phase II at a temperature of the release of H_2 and O_2, peak at 398.37 °C [20]. The first endothermic peak occurs in the temperature of 255.76 °C, a transition commonly called temperature decomposition which was consistent with literature values of 230 °C. The TG-DTA thermogram of carbon-polyvinyl alcohol (2.5 and 5% (wt)), showed the decrease of endothermic peak.

The shape of the mass loss curve for the inert atmosphere was consistent with the generally accepted two-step mechanism for the degradation coconut shell carbon-PVA and polyvinyl alcohol. The first step in the mass loss curve results from the elimination of water [18]. In the second step, in the inert atmosphere, pyrolysis occurs to produce some organic volatiles which were resulted in the second mass loss step [20-21]. The inert atmospheres of the material residues are respectively 93.8104, 10.5387 and 92.7250% this expected to be since pyrolisation results in the formation of amorphous carbon or semicrystallin carbon [22].

SEM-EDS analysis

Fig. 2 and 3 showed the SEM photographs of coconut shell surface carbonized at 1023 K and carbon composite (C/PVA) sintered at 1673 K. Fig. 2 showed that coconut shell carbon was non-homogeneous particles distribution. Fig. 3 showed a homogeneous and even surface morphology. The carbon composite from coconut shell carbon/polyvinyl alcohol (PVA) sintered at temperature of 1673 K exhibited uniform particle size of micrometer dimensions and spherical

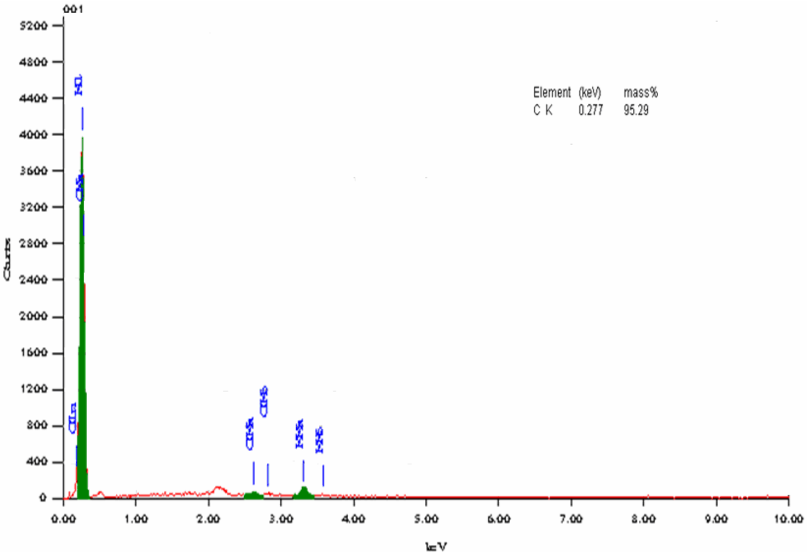

Fig 4. EDS analysis of the coconut shell carbon calcined at 1023 K, atmosphere Nitrogen

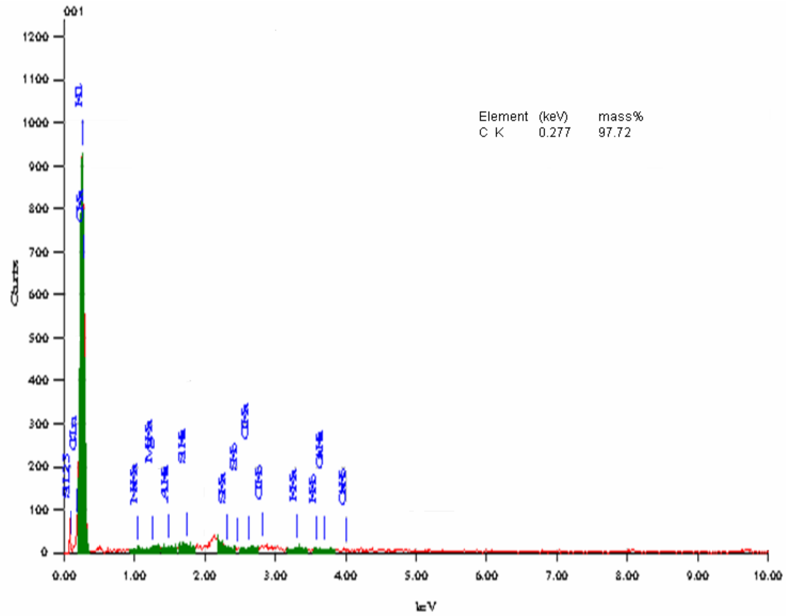

Fig 5. EDS analysis of the coconut shell carbon/PVA (5% (wt)) sintered 1673 K, atmosphere Argon

particles shape [1]. It was also shown in fig. 3 that the morphological features of carbon particles appeared to be more irregular, characteristics as a typical of carbon particles [2].

EDS analysis for both the coconut shell carbon calcined at 1023 K and carbon composite, carbon/PVA (5% (wt)) sintered at 1673 K were shown in Fig. 4 and 5. It was apparent that chemical composition of the coconut shell carbon (Fig. 4) consisted of C element up to 95.29% (wt) with average of C element was 94.97% (wt). The carbon composite, carbon/PVA (5% (wt)) (Fig. 5) consisted of higher C element up to 97.72% (wt) with average of C element was 97.44% (wt).

Heat treatment ranging from 1023 to 1673 K in this study, owing to the evolution of volatile matter [24] and the breakage of cross-linking bonds such as CH_2 bridge, the aromatic layers grow and coalesce each other [2].

FTIR Analysis

Fig. 6 showed that the three spectra between charcoal (raw materials), charcoal results in HCl washing, and calcination charcoal (1023 K, atmospheric nitrogen gas) that has been dried at a temperature of 393 K. There was no significant difference

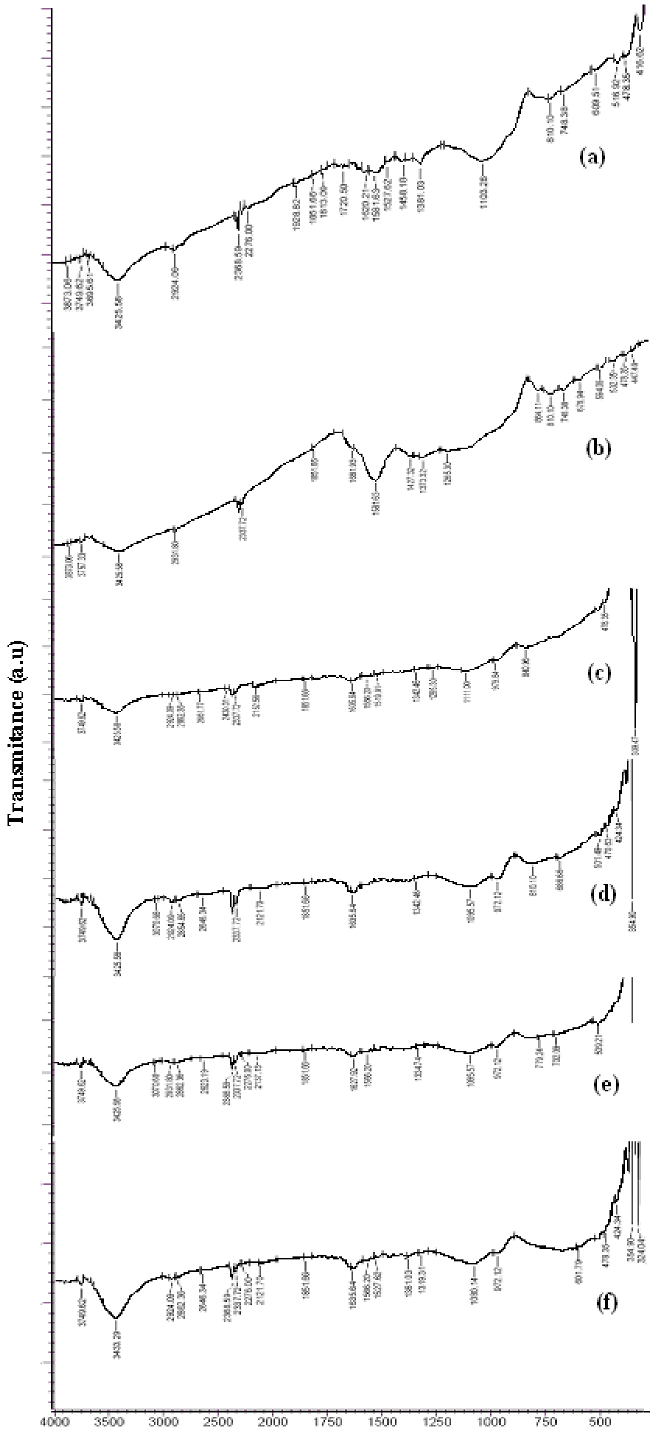

Fig 6. FTIR spectra pattern of carbon and PVA, solvent method, sintered at 1673 K, Argon atmosphere : (a) raw material of charcoal, (b) charcoal calcined at 1023 K, (c) raw material of carbon, (d) 2.5% (wt) PVA, (e) 5% (wt) PVA, and (f) 7.5% (wt) PVA

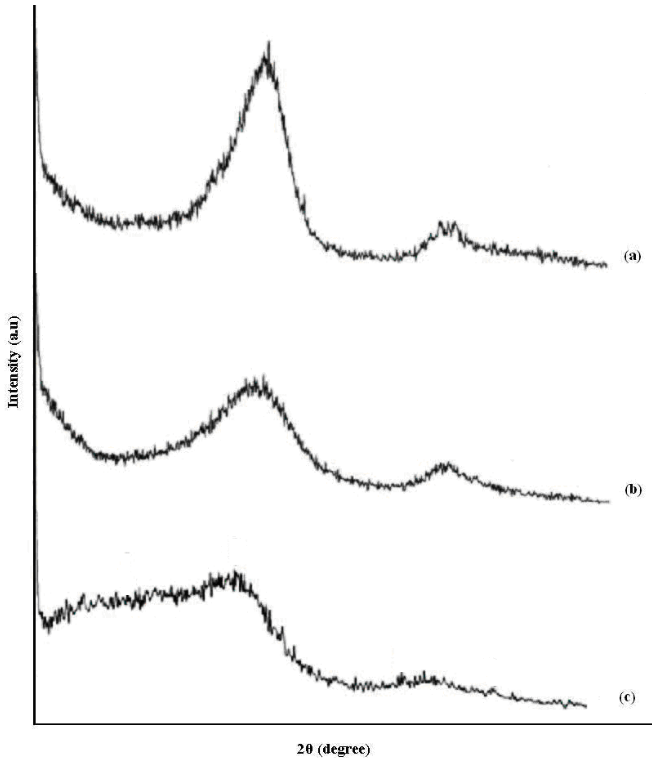

Fig 7. XRD pattern of : (a) coconut shell charcoal, (b) carbon calcined at 1023 K (N_2), and (c) carbon sintered at 1273 K (Ar)

(mean). All three spectra gave the peak absorption band that basically has the same pattern. Peak absorption bands that always exist continuously on the wave number of 3425.58 cm^{-1} showed stretching vibration of OH groups on the surface of the outer and inner bound to the elements of C contained in charcoal (such as carbon black) or can be water that is absorbed on charcoal [24-25]. Ribbon uptake 2931.50 cm^{-1}; 2337.72 cm^{-1} was a weak band, 1581.63 and 1103.28 cm^{-1} sharp and very strong characterizes the C-C stretching vibration found in the structure of carbon black.

FTIR spectra pattern of carbon composite produced by solvent method with PVA stimulant concentration of 2.5, 5 and 7.5% (wt) sintered at (1673 K, reaction time 3 h, argon atmosphere, annealing) was shown in Fig. 6 (a-d). The spectra showed a broad band near 3425 cm^{-1} which indicated the presence of hydroxyl groups on the carbon surface. The stretching was attributed to the absorbed water on the surface of coke and carbon black. The stretching frequencies of the aromatic C=C and aromatic C-H groups gave rise to peaks at 2924 and 2862, respectively, which were originally existed as the support for the coke and graphite [9,24]. The peak at near 1600 cm^{-1} ($\upsilon_{C=O}$) along with another peak at

1635 cm^{-1} (υ_{C-O}) indicated the presence of carboxylic acid groups. The peak at 1635 cm^{-1} was assigned to a conjugated hydrogen bonded carboxyl groups, assigned by several author on coke. The peak at 2924 cm^{-1} was due to C-H stretching of CH_2 groups [25]. The bands near 1600 cm^{-1} indicated the fingerprint region of C=O, C-O and C-H groups that exist as functional groups of coke and carbon black. The band near 972 cm^{-1} was due to out-of-plane bending of C-H group in benzene derivatives [18].

XRD analysis

Fig. 7 showed a typical X-ray diffraction (XRD) pattern of testing charcoal, calcined carbon and coke carbon material that sintered at temperatures 1273 K. XRD diffractogram of charcoal, calcined carbon and coke carbon material gave the value of d(Å) as a graphite semi-crystalline structure. Yin et al. [23] reported that the decrease of the interlayer spacing (d) and the increases of the crystalline diameter and average stacking height of the aromatic carbon sheets (L_c) with increasing the temperature suggested the development of stacking structure, increased the size of crystallite as well as removal of defects and increased order in carbon materials structure. The increase of L_c with increasing temperature resulted from crystallite growth in-plane and coalescence of crystallites. In this process, the structure of amorphous carbon structure was changed into semi-crystalline structure with a better degree of order, namely turbostatic structure [22]. There was the change of internal structure by setting the position of equilibrium carbon atoms.

CONCLUSION

The results showed that the products were in uniform particle sizes of micrometer dimensions and spherical particles shape, with average content of C element was 97.44% (wt), aromatic character and semi-crystalline structure.

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