



EFFECTS OF TEDA DOPING ON THE ADSORPTION PROPERTIES OF ACTIVATED CARBON AS THE FILLING MATERIAL OF IODINE SAMPLER FILTER ON THE KLK06 CR004 SYSTEM IN G.A. SIWABESSY MULTIPURPOSE REACTOR (RSG – GAS)

Cahyani Nelti Ulan^{*.1}, Vieska Rofianissa¹, Diene Noor Haerani², Nazly Kurniawan¹

¹Directory of Nuclear Facility Management - BRIN Kawasan puspitek Gd. 31, Setu, Tangerang Selatan, Indonesia

² Research Center for Nuclear Reactor Technology – BRIN, Kawasan Puspiptek Gd. 80, Setu, Tangerang Selatan, Indonesia

*Corresponding author
Email: cahy020@brin.go.id

Abstract. *The performance of KLK06 CR004 as the iodine sampler system is certainly related with the specification, also with the adsorption properties and capacity of the filter filling material, which is applied on that system. A certain Activated Carbon material is applied as the filling material for the filter part of KLK06 CR004, for the purpose of radioactive iodine sampling from the released air through the RSG – GAS stack. The characterization of the activated carbon for KLK06 CR004, with and without TEDA doping, has been carried out to develop as a candidate for the iodine sampler system. The material used in this research was The KLK06 CR004 activated carbon and The KLK06 CR004- TEDA doping with various composition of TEDA. The surface morphology of the activated carbon was investigated by Scanning Electron Microscope (SEM). Moreover, the qualitative and quantitative analysis of the activated carbon composition was also carried out in this research, by occupying the Energy Dispersive Spectroscopy (EDS). The adsorption surface area and particle size sample were analyzed by the Particle Size Analyzer (PSA) and Brunauer, Emmett, and Teller (BET).*

Keywords: *Iodine Sampler System; Adsorption Capacity; BET; PSA; SEM; and EDS.*

1. Introduction

Several studies regarding the capture of radioactive iodine compounds by applying the porous sorbents has resulted in significant interest over recent years as it is related to operational safety and the treatment and safe disposal of generated radioactive waste. When nuclear accident occurs, such as what happened in Chernobyl (1986) and Fukushima (2011), gaseous radionuclides have been released with significant quantity. The released radionuclides are mostly highly volatile and harmful long-lived ¹²⁹I, and also the short-lived ¹³¹I. [1] Iodine-137 has been by far the most serious pollutants in the general environment from fallout and as atomic wastes. Iodine-131 (half-life, 8 days) has been a serious pollutant in spite of its low half-life because it has been produced in high yield by fission (both in weapons and reactors). It is efficiently taken by cows from grass and hay and put into milk and one tiny gland, the thyroid of man and animals concentrates it very effectively

[2].

Activated carbon is often applied in nuclear technology as a filter in the emergency ventilation system of the reactor building which is switched on automatically when reactor failure occurs. Among the various fission product released during nuclear plant operations, radioiodine is considered as the most important because it produced radioactive iodine isotopes I^{131} and I^{133} . I^{131} is the most abundant isotope and combined with hydrocarbons to produce $CH_3 I^{131}$ which is highly poisonous. Activated carbon can be used for the removal of elemental radioactive iodine and its components [3]. However, the removal efficiency of the activated carbon is very small because of the propensity of water adsorption. Therefore, activated carbons are doped with different chemical reagents such as potassium iodide (KI) or Triethylenediamine (TEDA) [4]. One of the earliest compounds used for the treatment and capture iodine compounds are KI and TEDA. TEDA was selected for use as the dopant material in this research, and as it is known, the use of TEDA as a dopant on activated carbon originates from its application in the nuclear industry. There is an acknowledgement that the adsorption capacity of activated carbon for radioactive methyl iodide, the common form or compound of radioactive iodine, could be significantly increased by applying TEDA as the dopant material for the activated carbon. For increasing the iodine adsorption capacity of activated carbons, these materials are doped with TEDA with a concentration usually between 0 and 10wt% [5].

The doped TEDA activated carbon leads to an increase of the methyl iodide adsorption efficiency under high humidity conditions because it provides the solid a loftier functionality to adsorb the methyl iodide by the mechanism of specific dispersive interactions between methyl iodide and TEDA to form stable ammonium salts, which remain fixed within the adsorbent porous structure [6]. The general mechanism of that reaction is provided in Figure 1 [7,8].

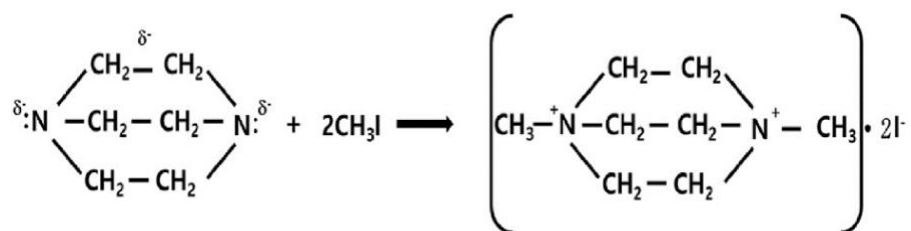


Figure 1. Chemical reaction between radioactive methyl iodide with the doped TEDA on the activated carbons.

Most researchers have suggested an adsorption mechanism consisting of two simultaneous dissimilar processes which are physical reversible adsorption and chemical irreversible adsorption. There is a clarification that the capture of radioactive methyl iodide by TEDA was a first-order catalytic reaction. However, another study justified that the doping of TEDA into activated carbons is transformed by reacting with iodine compounds such as methyl iodide *via* SN2 nucleophilic

substitution to become a quaternary amine [9].

Activated Carbon has been used as the main material of the iodine sampler system. This material has excellent properties in adsorption of radioactive iodine sampling from the released air through the RSG – GAS stack. Iodine adsorption system in RSG – GAS is usually performed by KLK06 CR004. This system is able to collect iodine in any form such as elemental iodine and methyl iodide [10]. The air to be measured passes through the iodine filter cartridge. This cartridge is filled by activated carbon material, as the iodine collector. During the operation period of RSG – GAS, the KLK06 CR004 system is always activated as the iodine collector. After the reactor operation, the iodine concentration in the activated carbon inside the iodine filter cartridge is investigated by a gamma spectrometry system. Therefore, the effectivity of activated carbon for adsorbing the iodine is an important thing [3].

The information about technical specification of the activated carbon; which describes the adsorption properties and the adsorption capacity of the activated carbon, is still not available up to now. In this research; samples of the activated carbon from KLK06 CR004 system were doped by applying TEDA. Then, the characterization of the TEDA doped activated carbon samples was carried out by occupying the Brunauer, Emmett, and Teller (BET) Surface Area Analyzer, the Scanning Electron Microscope (SEM), and the Energy Dispersive Spectroscopy (EDS). Those treatment and investigation are carried out in order to figure out the adsorption properties and the adsorption capacity of the KLK06 CR004 activated carbon; after the modification of the previous activated carbon, which is done by applying the TEDA as the dopant material.

Activated carbons are well known for their high surface area and complex pore structure. Since it is possible to prepare a wide range of activated carbons from many precursors using different activation methods, it is important to have a method of characterizing the properties of these materials. Until now, the most widely accepted method for such characterization is the measurement of adsorption and desorption isotherms of probe molecules with known molecular dimensions. Theoretical models for the adsorption of gas molecules onto porous surfaces combined with experimental data allow elucidation of properties such as apparent surface area, pore size distribution and pore volume. The most popular adsorbate molecule for this purpose is nitrogen due to its non-polar, inert nature and small size. The BET surface area analyzer operation is based on that concept. The presence of dopant material, such as TEDA, will probably reduce the surface area of the activated carbon. However, proportional concentration of TEDA will increase the iodine adsorption capacity of the activated carbon, due to the occurrence of reactions between the dopant and the contaminant to yield quaternary ammonium salts [6].

Moreover, the surface morphology and the porosity structure of the activated carbon is investigated by performing the SEM observation. The SEM utilizes a beam of electrons to scan

the surface of a material and has the capability to perform the direct observation of material surface characteristics at the very micro levels. Due to the great range of its magnifications and sharp resolutions which can be obtained with the SEM, it has been applied in a lot areas of science and industry, especially in the fields of materials engineering, biological and medical sciences for the investigation and characterization of the micro-structure of materials.

The SEM generates a focused beam of electrons to make the detailed image of a material and to obtain information about its structure and composition. The SEM is capable of providing the information about the topography, morphology, composition, and crystallographic information [11]. With the SEM characterization capability, then, there is a lot of opportunity to resolve the previous unaddressed issues regarding the characterization development of the activated carbon porosity. This capability also makes the SEM to become a very useful instrument for the advanced study of the activated carbons.

The topographic information possibly gained using the SEM allows that surface details such as pore characteristics, the description of which has been a major focus of activated carbon researcher, to be investigated and analyzed directly from SEM micrographs. Also, the ability of SEM to disclose compositional details of a sample makes it a reliable instrument for researching the surface chemistry, also the characteristics of the activated carbons. Additional features of the SEM make it capable of disclosing the details of a sample with the size less than 1 nm. The SEM images of the activated carbon, without and with the presence of dopant material, are as shown in Figure 2 [12].

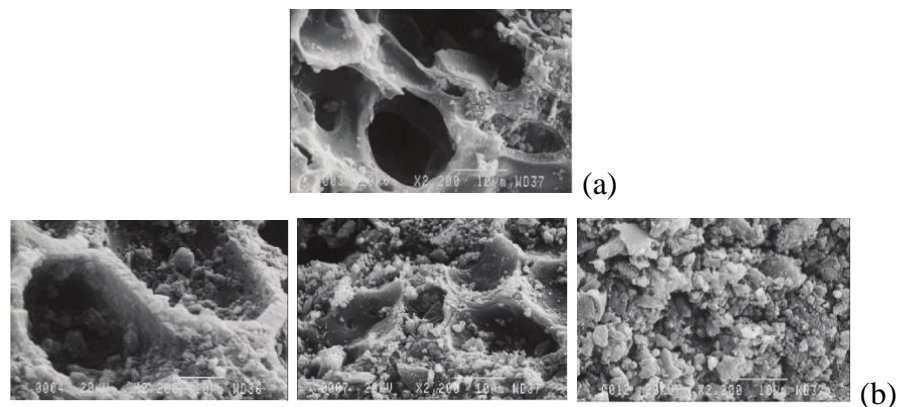


Figure 2. SEM images of undoped activated carbon (a), and doped activated carbon with different concentration of the dopant (b) [12]

Energy Dispersive Spectroscopy (EDS) analysis was employed to observe the physical morphology of sample surfaces and to analyze the element compositions, including the visible light elements such as carbon, nitrogen and oxygen. EDS detector equipped with ultra-thin element light windows that detect elements with atomic numbers > 4 . Qualitative elemental analysis involves the assignment of elements to the characteristic X-ray peaks recognized in the EDS

spectrum [13]. This function is routinely performed with automatic peak identification (e.g., “AutoPeakID”) software embedded in the vendor EDS system. As characteristic X-ray intensities are proportional to the particular element’s concentration, therefore, semi-quantitative analysis can be performed. The concentrations of unknown elements in a sample can be obtained by matching the intensities of characteristic X-ray of standard sample with that of the unknown sample. However, X-Ray generated in the sample may be either absorbed within the sample or excite the X-rays from other elements before being emitted in vacuum. Therefore, correction is required in this analysis method for which the several requirements must be considered.

By several studies of this research, it is expected to obtain the information about the optimum concentration of the TEDA, which can be applied as the dopant material for the modified activated carbon of the KLK06 CR004 system. With the optimum concentration of the TEDA for modifying the previous activated carbon [6, 14], it will obtain the more efficient of iodine detection during the RSG – GAS operation. The characterization studies of the modified activated carbon are also expected to provide useful information about the properties of the modified activated carbon, which implies or indicates the iodine adsorption capacity of the modified activated carbon.

2. Methods

2.1. Material

In this research, the activated carbon from the cartridge of KLK06 CR004 was taken as the granular carbon samples, for the synthesis of composite or modified activated carbon, which was referred as the TEDA doped activated carbon. The chemicals used in the synthesis of composite or modified activated carbon include: triethylenediamine (TEDA), ethanol 96 % (specification), NaOH (specification) and aquadest.

The equipment used in this research includes: High Energy Milling (HEM), Hot Plate with Magnetic Stirrer, Furnace, Filter Paper, Laboratory Glassware, Brunauer, Emmett, and Teller (BET) Surface Area Analyzer, Scanning Electron Microscope (SEM) – Energy Dispersive Spectroscopy (EDS), and other required tools, detailed according to the implementation stage. The stages in which the tools used are as follows:

1. Synthesis of Composite or Modified Activated Carbon

Equipment for the synthesis of composite or modified activated carbon: analytical balances, high energy milling (HEM), hot plate with magnetic stirrer, furnace, oven, filter paper, and glassware commonly available in laboratories.

2. Characterization of The Activated Carbon

Equipment for characterization of the activated carbon: Brunauer, Emmett, and Teller (BET) surface area analyzer, scanning electron microscope (SEM) – energy dispersive

spectroscopy (EDS), analytical balances, oven, porcelain dishes, desiccators, furnaces, and glassware commonly available in laboratories.

2.2. Methods

A. Preparation

This research stages began with the size reduction of the granular carbon, to obtain powder sized carbon, using the High Energy Milling (HEM).

B. Synthesis of Composite or Modified Activated Carbon

1. The activated carbon samples were made into 4 different types of samples, with the weight of each sample was 5 grams:
 - a. Activated carbon without TEDA
 - b. Activated carbon with 0.15 grams of TEDA (3%)
 - c. Activated carbon with 0.30 grams of TEDA (6%)
 - d. Activated carbon with 0.45 grams of TEDA (9%)
2. Then, each sample was dissolved by using 8 ml of ethanol 96%. Each mixture was stirred and heated at 76°C, for 2 hours.
3. The 22.5 mL of 2 M NaOH solution was added into each mixture, and each mixture was stirred for 1 hour.
4. Each mixture, then, was allowed to stand for 12 hours. After that, each mixture was filtered by using filter paper. The sediment obtained from the filtration process, then, was dried in the oven at 60°C.

C. Characterization of The Activated Carbon

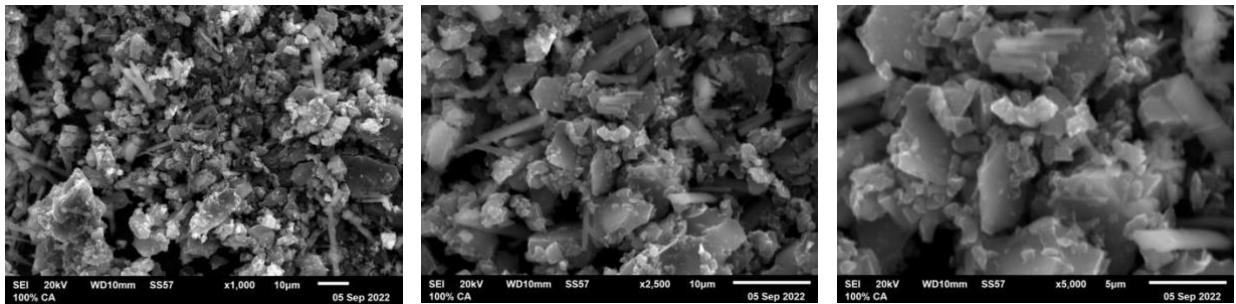
The surface area of each sample was measured by occupying the Brunauer, Emmett, and Teller (BET) surface area analyzer. Afterwards, the surface morphology, also the qualitative and quantitative of each sample was observed by occupying the scanning electron microscope (SEM) – energy dispersive spectroscopy (EDS).

3. Results and Discussion

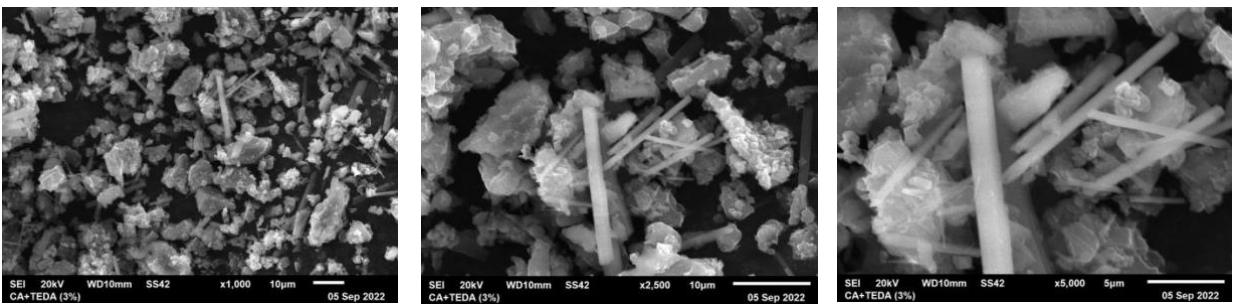
3.1. Surface morphology analysis

The surface morphology of the activated carbon (AC) sample without TEDA dopant (0%) also the activated carbon with TEDA (3%, 6%, dan 9%), has been investigated using the Scanning Electron Microscope (SEM); with the magnification up to 5000 times, as shown in [Figure 3](#). The SEM image, resulting from the secondary electron (SE) imaging; shows that, the activated carbon samples have heterogenous form, which are in the form of flake, granule, and rod. These heterogenous form is possibly formed by the constituent raw materials of the activated carbon, and due to this condition; the activated carbon becomes a porous material, which is formed by the

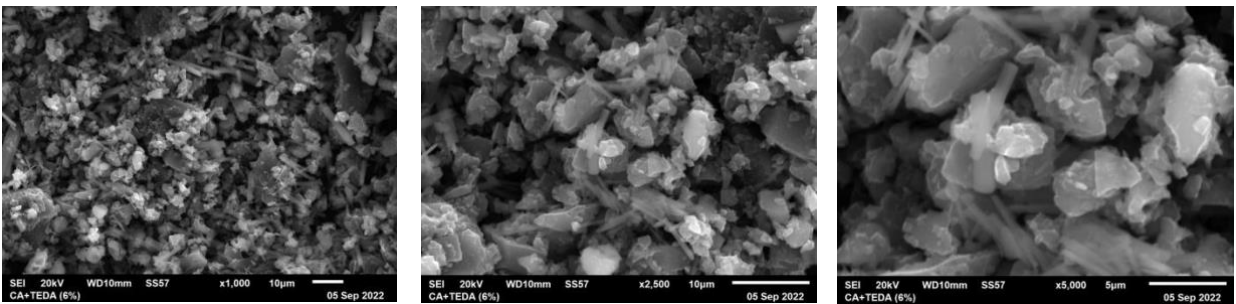
combination of these form in the solid phase. Also, due to these heterogenous form; each activated carbon sample has a wide range of its particle size, which may vary from 1 μm up to 10 μm .



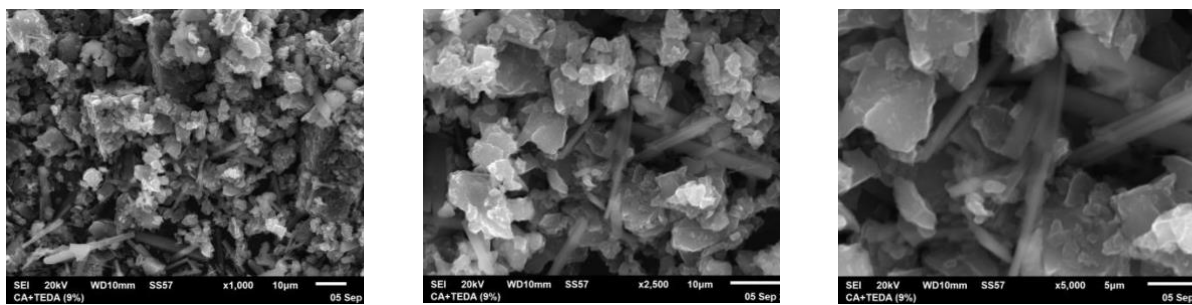
(a) AC without TEDA (0%)



(b) AC with TEDA (3%)



(c) AC with TEDA (6%)



(d) AC with TEDA (9%)

Figure 3. The surface morphology of the activated carbon samples, investigated by SEM, with magnification from 1000, 2000, and 5000 times.

Whereas, the activated carbon sample with TEDA (3%, 6%, and 9%) has similar surface morphology with the pure activated carbon, since TEDA does not affect the microstructure

physically, rather attached to the surface layer and the pore of the activated carbon to increase the iodine adsorption capacity by the reaction of the attached TEDA and iodine [15, 16]. Each sample has been investigated with the magnification of 1000, 2500, and up to 5000 times; thereby, it can be disclosed that the three types of the particle form are located at the whole surface of the sample. This morphology is partly similar with the identification results which are provided by Maghsoud Gourani dan Matthew Chinn [12, 15]. They successfully obtained the surface features of the activated carbon such as the pore characteristics. Further investigation for qualitative and quantitative analysis of the activated carbon is possible to be carried out using back scatter electron (BSE) method of the energy dispersive spectroscopy (EDS) analysis, which is coupled with the SEM [13]. The nature of activated carbon is highly dependent on the activation process and the nature of the base material so that it greatly affects the change in adsorption capacity, porosity, surface area, and pore volume [3, 17, 18]. The heterogenous form of the activated carbon samples lead to the explanation that they have a relatively high total surface area with a relatively high level of porosity and also geometrically solid.

3.2. Energy Dispersive Spectroscopy Analysis

In this research, the energy dispersive spectroscopy (EDS) is applied to figure out the element composition in each sample. Before conducting the investigation and the analysis, for the activated carbon (AC) sample without TEDA dopant (0% TEDA); also, for the TEDA doped activated carbon samples (with 3%, 6%, and 9% of TEDA), each sample was dissolved into 96% ethanol, and 2M NaOH was added into each solution. Based on the semi-quantitative analysis result of each sample, the congruence of stoichiometry composition of each sample is able to be confirmed. Figure 4 shows the analysis results regarding the elementary composition of the sample without TEDA dopant and the TEDA doped samples.

Figure 4 shows that several elements in the activated carbon sample without TEDA dopant are detected. Carbon (C) at energy channel of 0.277 keV, oxygen (O) at 0.525 keV, and natrium (Na) at 1.041 keV. The detected C and O are possibly the constituent of the activated carbon and ethanol. Meanwhile, the Na is possibly the constituent of NaOH. However, the very light element; H, was not able to be detected by EDS analysis.

Whereas, in the analysis results of the TEDA doped activated carbon (3%, 6%, and 9%); besides the constituent of activated carbon itself and the constituent of ethanol, the detected C also comes from the hydro-carbon group of the TEDA. However, there is an anomaly regarding the analysis result of the activated carbon sample with 9% TEDA since the dispersive energy peak of calcium (Ca) also appears. Yet, still the source of Ca remains unknown. Then, a normalization toward the sample is required for eliminating the Ca. Moreover, the semi-quantitative analysis result, regarding the elements content, is as shown in Table 1.

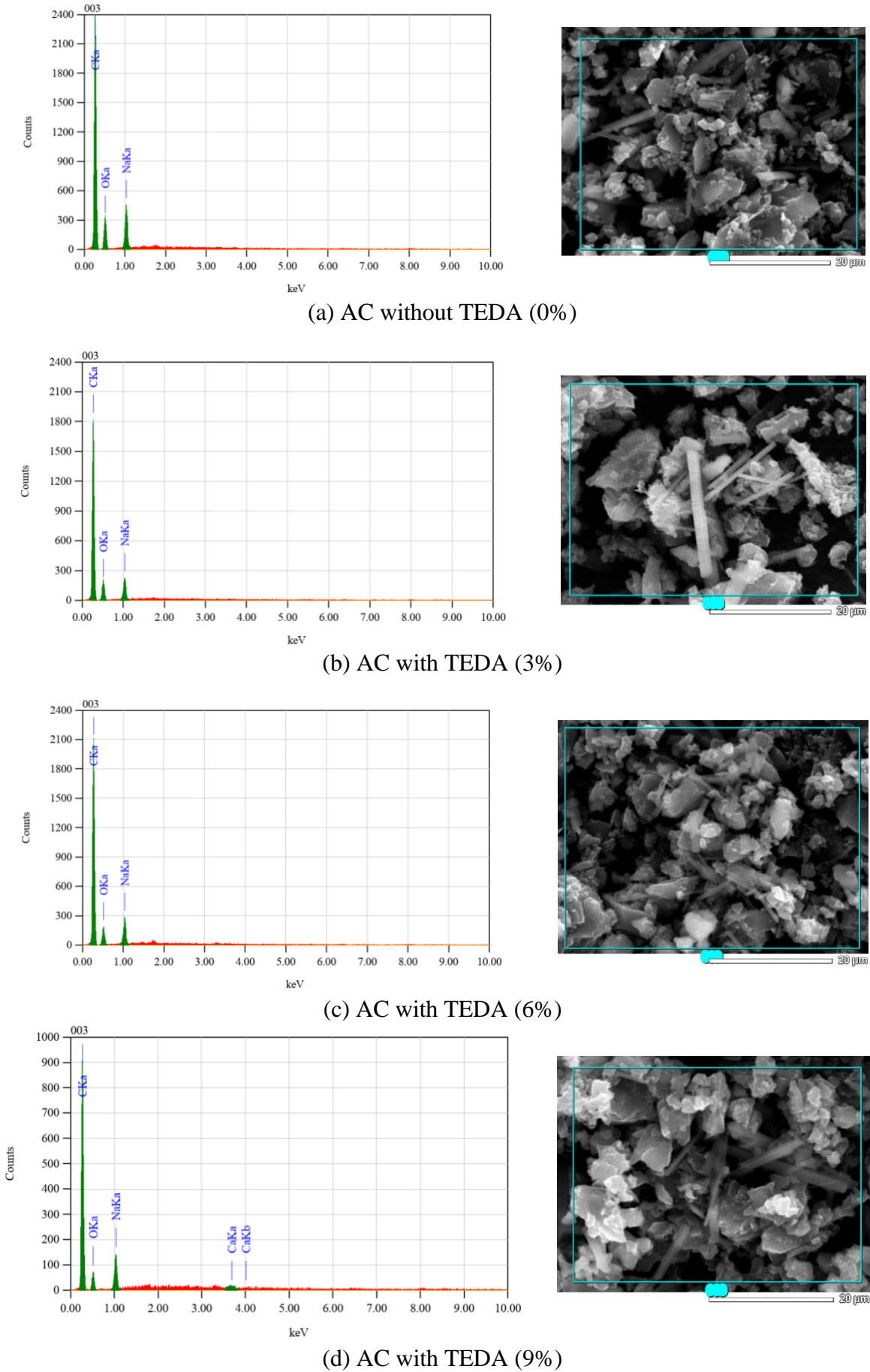


Figure 4. The EDS analysis results by applying the energy channel from 0 up to 10 keV.

Table 1. The EDS analysis result regarding the elements content of the activated carbon samples

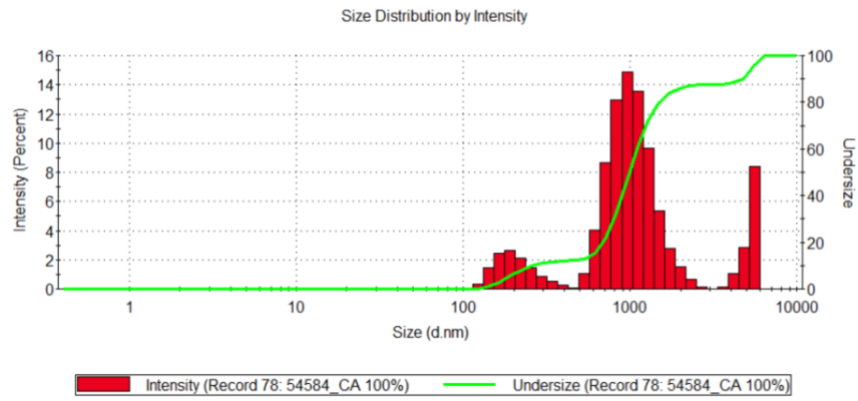
Nr.	Elements	Content (wt %)			
		AC without TEDA	AC with 3% TEDA	AC with 6% TEDA	AC with 9% TEDA
1.	Carbon (C)	68.14 ± 0.04	70.32 ± 0.04	73.10 ± 0.04	74.21 ± 0.05
2.	Oxygen (O)	25.45 ± 0.41	24.68 ± 0.36	21.35 ± 0.41	20.03 ± 0.51
3.	Sodium (Na)	6.41 ± 0.13	5.00 ± 0.11	5.54 ± 0.12	5.76 ± 0.14

Table 1 shows that the C content increases with the concentration increase of the doped TEDA, from 68.14 wt% up to 74.21 wt%. This means that the presence of C element is not only from the activated carbon and ethanol, as the activating reagent. There is also C element contribution from hydro-carbon group of TEDA, as the dopant material. The increase of TEDA content in the activated carbon samples may prove that the doping mechanism of TEDA to the activated carbon samples has met the expectation.

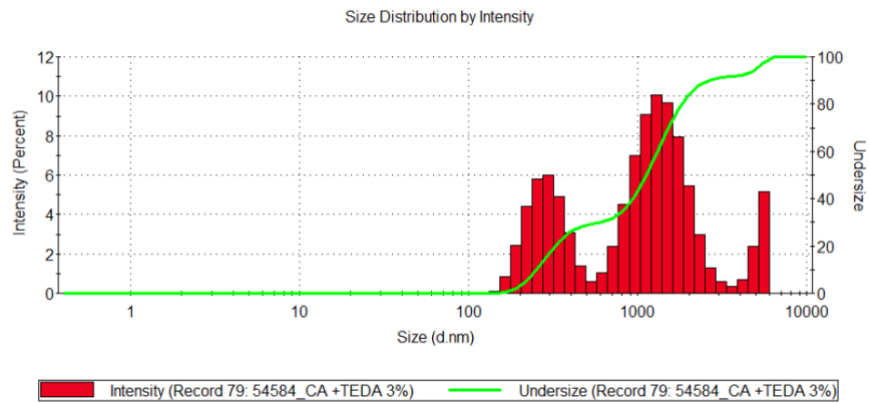
As the dopant materials in the activated carbon samples, the presence of TEDA will also change the content value of O and Na. The content value of O will decrease with the presence of TEDA, and it will be quite decreased with the increase of the doped TEDA concentration. Meanwhile, the content value of Na will significantly decrease after the TEDA doping. However, the content of Na will be slightly changed with the increase of the doped TEDA concentration. Therefore, according to the result of this elementary analysis; the increase of the C content by the concentration increases of the doped TEDA, has provide an indication; that the TEDA has successfully been doped to the activated carbon samples by applying the method in this research.

3.3. Particle Size Analysis

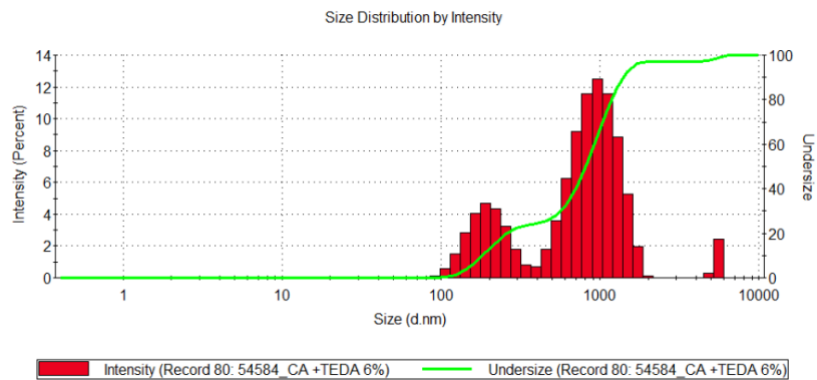
The analysis regarding the distribution of particle size was carried out by Particle Size Analyzer (PSA) using Dynamic Light Scattering (DLS) method by Non-Invasive Back Scatter (NIBS) technique. The sample was tested in a 4 mW He-Ne laser source with a wavelength of 633 nm, with measurement range (hydrodynamic size) of 0.6 nm – 9000 nm [19]. The sample has to be in liquid form, or nanometer-sized powder dispersed in a liquid medium. Particle size can calculate the particle size from the angle of the light scattered by the particle stream passing through the laser beam. DLS can determine the size of the intensity fluctuations of the scattered laser beam created by the Brownian motion of the particles. The induced lattice particle size analyzer identifies the smaller particle sizes in solution by electrically aligning the particles and then measuring their diffusion. Figure 5 shows the results of the particle size analyzer (PSA) analysis of activated carbon without TEDA (0%) and activated carbon (AC) with TEDA (3%, 6%, and 9%).



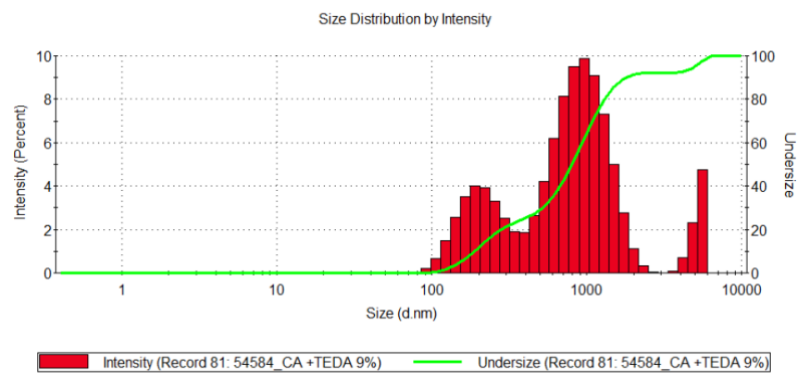
(a) AC without TEDA (0%)



(b) AC with TEDA (3%)



(c) AC with TEDA (6%)



(d) AC with TEDA (9%)

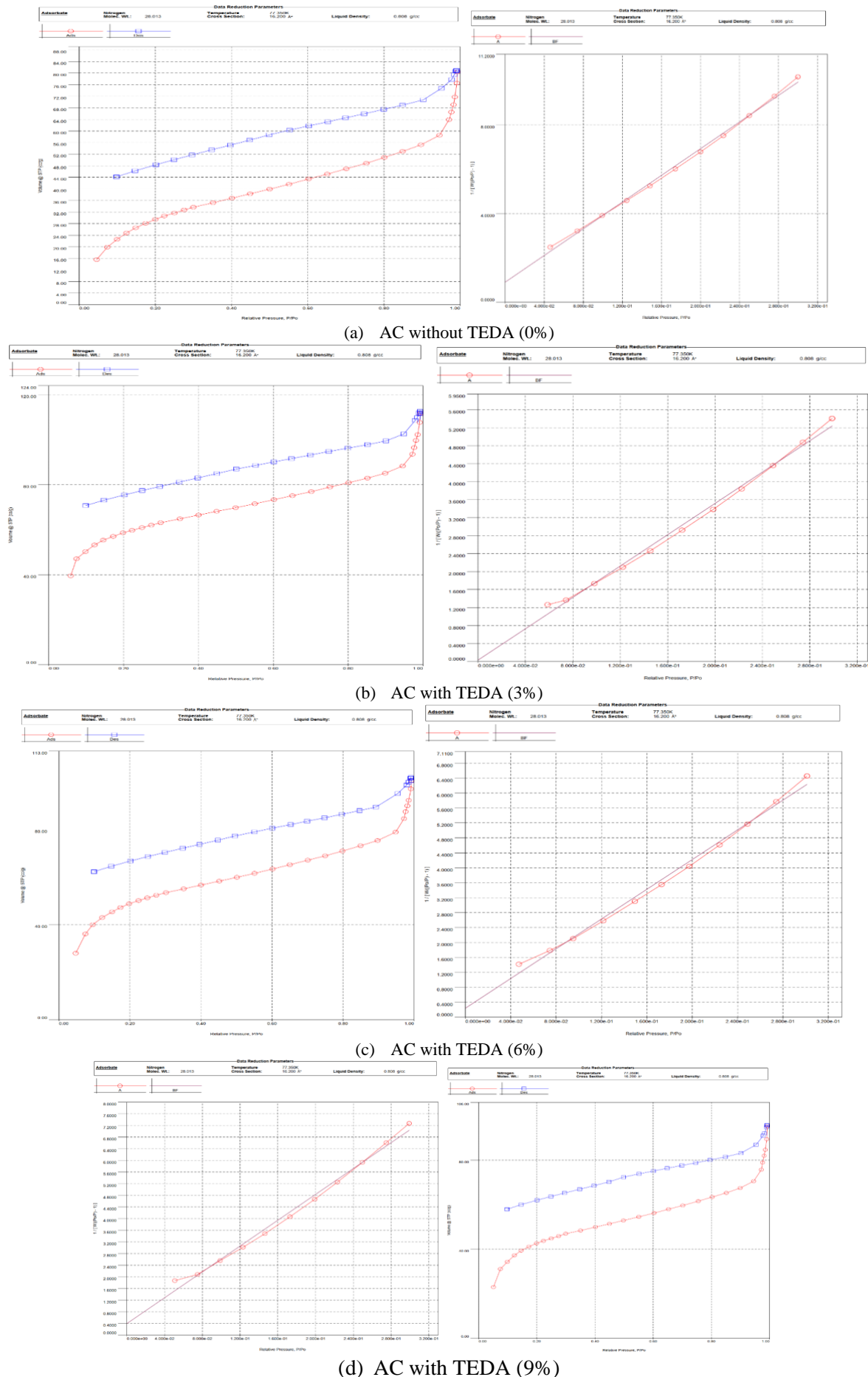
Figure 5. The results of the particle size analysis of the four samples using PSA measured from the range of 0.6 nm – 9,000 nm.

The results of PSA analysis are used to provide confirmation and supporting data based on the results of particle morphology observations using Scanning Electron Microscope (SEM). It was stated that based on the results of SEM photos from imaging using secondary electrons (SE), it appears that both the activated carbon without TEDA sample and the activated carbon with TEDA sample (3, 6, and 9%) have similar and heterogeneous shapes including flakes, granules, and rods.

This statement is confirmed by the PSA measurement data which shows that the active carbon sample without TEDA has 3 types of particle sizes which are indicated by the presence of 3 peaks on the PSA curve. This means that the three peaks indicate that the sample consists of 3 different particle size ranges. The small, medium and large particle size ranges are similar to the results of observations with SEM where the small particle size range is thought to be in the form of granules, the medium particle size is thought to be in the form of small flakes, and large particle sizes are assumed to be rods and large flakes. The activated carbon sample without TEDA has 3 particle sizes in the range of 100 nm to 500 nm, 500 nm to 1200 nm, and above 1200 nm, which are 12.4, 75.3, and 12.3%, respectively. The activated carbon sample with TEDA 3% also has 3 particle sizes in the range of 100 nm to 500 nm, 500 nm to 1300 nm, and above 1300 nm, which are 29.2, 62.3, and 8.5%, respectively. The activated carbon sample with TEDA 6% also has 3 particle sizes with a range of 100 nm to 500 nm, 500 nm to 1100 nm, and above 1300 nm sizes which are 24.4, 72.8, and 2.7%, respectively. While the activated carbon sample with TEDA 9% also has 3 particle sizes with a range of 100 nm to 500 nm, 500 nm to 1300 nm, and above 1300 nm size which are 25.5, 66.8, and 7.7%, respectively. It is clear that TEDA does not physically affect the microstructure, but only absorbs the sample which can increase its adsorption capacity.

3.4. Adsorption-Desorption and Pore Analysis

In [Figure 6](#), it is shown the curve hysteresis isotherm adsorption-desorption Nitrogen (N_2) at temperatures of 77 k from samples activated carbon (AC) without TEDA and activated carbon with TEDA (3, 6, and 9 %). An analysis in the form of a curve hysteresis isotherm adsorption describes the occurrence of interaction between N_2 gas with a solid activated carbon that also describes the system geometry pores in sample. The four samples both activated carbon without TEDA, activated carbon with TEDA (3, 6, and 9%) show a relatively small hysteresis curve widening and adsorption properties are still limited to high P/Po. This means that the four samples of activated carbon have relatively low mesoporous system. However, by increasing the content of TEDA, the isotherm adsorption is able to be increased. The widening of hysteresis curve shows that the material is mesoporous due to capillary condensation in the mesopore system.



(a) AC without TEDA (0%)

(b) AC with TEDA (3%)

(c) AC with TEDA (6%)

(d) AC with TEDA (9%)

Figure 6. Hysteresis curve of N₂ adsorption-desorption Isotherm at 77 K and Total surface area of all four samples.

Figure 6 shows also the measurement results of the total surface area of the material, which is analyzed based on the method of Brunauer, Emmet, and Teller (BET) [20]. Moreover, based on this BET analysis, pore size distribution was also obtained, where in general pore size can be classified into 3 types, which are; pore diameter size < 2 nm, pore diameter size between 2 nm up to 50 nm, and pore diameter size > 50 nm. These 3 types of pore size respectively referred to as micropores, mesopores, and macropores.

The measurement results of the total surface area of activated carbon samples without TEDA are $199.533 \text{ m}^2/\text{g}$. However, after adding TEDA with the concentration of 3%, 6%, and 9%, the total surface area decreased to $172.997 \text{ m}^2/\text{g}$, $154.102 \text{ m}^2/\text{g}$, and $112.168 \text{ m}^2/\text{g}$. The micropore was formed in the internal region and the mesopore was relatively developed near the surface region of the TEDA-AC. The specific surface area of the activated carbon was reduced by TEDA doping on its surface, and it is because micropore contributed more to form the specific surface area than the mesopore.

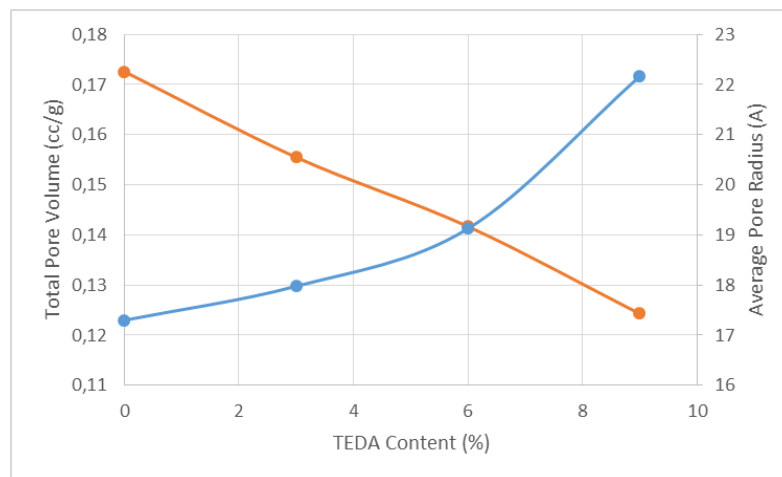


Figure 7. Total pore volume and average pore radius, as a function of increasing TEDA content.

Figure 7 shows the measurement results of total pore volume and pore radius average to activated carbon without TEDA and activated carbon samples with TEDA (3%, 6%, and 9%). Samples of activated carbon without TEDA have a total pore volume of 0.1726 cc/g with an average pore radius of 17.2993 \AA , while samples of activated carbon with TEDA (3%, 6%, and 9%) respectively have a total pore volume of 0.1555 cc/g , 0.1474 cc/g , and 0.1243 cc/g with the average pore radius is increased to be 17.9776 \AA , 19.1258 \AA , and 22.1643 \AA . The decrease in total pore volume is due to the doping of TEDA, which causing the TEDA take places in the pores of the activated carbon, and; furthermore, it also enlarges the average pore radius of activated carbon.

According to the pore size distribution of activated carbon without TEDA samples and activated carbon with TEDA samples (3%, 6%, and 9%), it appears that these four samples of activated carbon have the pore sizes, which can be categorized as the micropore and mesopore. Then, another obtained information from the analysis; is that, the initial micropore volume of the

activated carbon without TEDA and activated carbon with TEDA are respectively 0.086 cm³/g, 0.071 cm³/g, 0.062 cm³/g, and 0.041 cm³/g. Meanwhile, the mesopore volume are respectively 0.089 cm³/g, 0.087 cm³/g, 0.88 cm³/g, and 0.086 cm³/g. Two well-known analysis methods, such as the Horváth-Kawazoe (HK) method and Barrett-Joyner-Halenda (BJH) method, can be applied for obtaining these pore size values [20]. Next explanation, is that; the decrease of micropores and mesopores volume of the TEDA-AC is due to the doped TEDA takes place at the aperture of the micropores, and is also deposited on the mesopores; causing clogging of the mesopores and micropores, and therefore, decreasing the surface area of the TEDA doped activated carbon.

4. Conclusions

Activated carbon without TEDA sample and the activated carbon with TEDA sample (3%, 6%, and 9%) were analyzed by PSA, BET, and SEM-EDS. Based on the results of SEM and PSA, it appears that both the activated carbon without TEDA sample and the activated carbon with TEDA sample (3%, 6%, and 9%) have similar and heterogeneous shapes including flakes, granules, and stems. Based on the results of BET, the total surface area of activated carbon samples without TEDA is 199.533 m²/g. However, after adding TEDA with the concentration of 3%, 6%, and 9%, the total surface areas are respectively decreased to 172.997 m²/g, 154.102 m²/g, and 112.168 m²/g.

It also has been figured out, that; samples of activated carbon without TEDA have a total pore volume of 0.1726 cc/g, with an average pore radius of 17.2993 Å. Meanwhile, samples of activated carbon with TEDA (3%, 6%, and 9%) respectively have a total pore volume of 0.1555 cc/g, 0.1474 cc/g, and 0.1243 cc/g, with an average pore radius is increased to be 17.9776 Å, 19.1258 Å, and 22.1643 Å. The decrease in total pore volume decreased is due to the doping of TEDA, which causing the TEDA takes places in the pores of the activated carbon, and; furthermore, it also enlarges the average pore radius of activated carbon.

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References

- [1] E. Z. Ebeid., and M. B. Zakaria. Thermal Analysis: From Introductory Fundamentals to Advanced Applications. Netherland: Elsevier Science, 2021.
- [2] Grimes, Russell N., Cotton, F. Albert., Wilkinson, Geoffrey., Bochmann, Senior Lecturer School of Chemical Sciences Manfred., Bochmann, Manfred., Murillo, Carlos A.. Advanced

Inorganic Chemistry. Germany: Wiley, 1999.

- [3] J. Zhou., S. Hao., L. Gao., and Y. Zhang. "Study on adsorption performance of coal based activated carbon to radioactive iodine and stable iodine," *Annals of Nuclear Energy*, vol. 72, pp. 237–241, 2014, <http://dx.doi.org/10.1016/j.anucene.2014.05.028>
- [4] Novel Carbon Adsorbents. Netherland: Elsevier Science, 2012.
- [5] Sustainable Energy and Environmental Technology - Proceedings Of The Asia-pacific Conference. Singapore: World Scientific Publishing Company, 1996.
- [6] C. M. Gonz alez-Garc a., J. F. Gonz alez., and S. Rom n. "Removal efficiency of radioactive methyl iodide on TEDA-impregnated activated carbons," *Fuel Process. Technol.*, vol. 92, no. 2. pp. 247-252, 2011, <https://doi.org/10.1016/j.fuproc.2010.04.014>
- [7] C. Byung-Seon., K. Seon-Byeong, M. Jeikwon, S. Bum-Kyung, "Removal efficiency of radioactive methyl iodide on TEDA-impregnated activated carbons," *Nuclear Engineering and Technology*, vol. 53, no. 5, pp. 1519-1523, 2021, <https://doi.org/10.1016/j.net.2020.10.020>
- [8] G. I. Park, I. T. Kim, J. K. Lee, S. K. Ryu., and J. H. Kim. "Effect of Temperature on the Adsorption and Desorption Characteristics of Methyl Iodide over TEDA Impregnated Activated Carbon," *Carbon letters*, vol. 2, no. 1, pp. 9-14, 2001. <https://koreascience.kr/article/JAKO200122463504798.page>
- [9] J. Huve., A. Ryzhikov., H. Nouali., V. Lalia., G. Aug e., and T. J. Daou. "Porous sorbents for the capture of radioactive iodine compounds: a review," *RSC Adv.*, vol. 8, pp. 29248–29273, 2018, <https://doi.org/10.1039/c8ra04775h>
- [10] Editor Team of Safety Analysis Report, "Operational of Radiological Safety," in Safety Analysis Report of G.A. Siwabessy Multipurpose Reactor, Rev.11, National Nuclear Energy Agency, 2020.
- [11] O. W. Achaw, "A Study of the Porosity of Activated Carbons Using the Scanning Electron Microscope," in Scanning Electron Microscopy, Viacheslav Kazmiruk, IntechOpen, 2012, pp. 473-490.
- [12] M. Gourani., A. Sadighzadeh., and F. Mizani, "Effect of impregnating materials in activated carbon on Iodine - 131 (¹³¹I) removal efficiency," *Radiation Protection and Environment*, vol. 37, no. 3 & 4, pp. 179–183, 2014. <https://doi.org/10.4103/0972-0464.154882>
- [13] J. I. Goldstein, D. E. Newbury, J. R. Michael, N. W. M. Ritchie, J. H. J. Scott, and D. C. Joy, "Scanning Electron Microscopy and X-Ray Microanalysis, 4th ed." New York: Springer, 2018.
- [14] H. C. Lee., D. Y. Lee., H. S. Kim and C. R. Kim, "Performance evaluation of TEDA impregnated activated carbon under long term operation simulated NPP operating condition." *Nuclear Engineering and Technology* (2020), <https://doi.org/10.1016/j.net.2020.04.020>
- [15] Matthew Chinn, "The Impregnation of Activated Carbon," Ph.D. thesis, The University of Huddersfield, 2001.
- [16] A. Farooq, "Removal of Hazardous Pollutants by Impregnation and Parametric Optimization of Adsorbents," Ph.D. thesis, Dept. Chem. Eng., Pakistan Institute of Engineering and Applied Science, Islamabad, 2010.
- [17] K. Kaneko, "Determination of pore size and pore size distribution: Adsorbents and catalysts". *J. Memb. Sci.* 96, pp. 59-89, (1994).
- [18] J. Treese, C. Pasel, M. Luckas, and D. Bathen. "Chemical Surface Characterization of Activated Carbons by Adsorption Excess of Probe Molecules." *Chem. Eng. Technol.*, vol. 39,

no. 6, pp. 1144-1150, 2016.

- [19] T. Allen, Particle Size Measurement, Chapman & Hall. 4th Edition, 1992. ISBN 04123570.
- [20] M. Thommes, K. Kaneko, A. V. Neimark, J. P. Olivier, F. Rodriguez-Reinoso, J. Rouquerol., and K. S. Sing, “Physisorption of Gases, with Special Reference to the Evaluation of SurfaceArea and Pore Size Distribution (IUPAC Technical Report).” Pure Appl. Chem., vol. 87, no. 9-10, pp. 1051-1069, 2015. <https://doi.org/10.1515/pac-2014-1117>