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Preparation of Black Carbon Nanoparticles from Two Sources of Oil Residues with Direct and Indirect Ultrasonic Process

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ABSTRACT

This work compares two methods of producing carbon nanoparticles from oil waste or by-products that accumulate on the walls of pipes and furnaces and reduce the efficiency and life of the equipment (which can hinder the production process; therefore, it is removed weekly). These wastes or by-products are generated from the incomplete combustion of hydrocarbon compounds during the crude oil refining process to produce petroleum products. The raw materials were collected from two Al-Dura refinery sites, sieved, burned in a furnace under vacuum and inert atmosphere, crushed with a mortar and washed with solvent, washed with distilled water, dried, and then reduced to nanoparticle size by direct crushing (sonication with a probe) and indirect ultrasonic methods (bath sonication). The samples were analyzed after crushing and firing using the appropriate methods such as EDX and SEM as well as tests. Zeta potential and particle size analysis were two other tests performed on the final products. The results showed that the carbon content increased consecutively from 28.49, 36.30 to 91.59 and 94.47% after firing. In addition, the direct ultrasonic method is superior to the indirect method for producing carbon nanoparticles because it requires less time and can produce nanoparticles with an average grain size of about 37 nm and 86.6 nm for the first and second samples, respectively. The zeta potential data show that the resulting nanocarbon particles are relatively stable.

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1. Introduction

The process of refining crude oil is fundamentally conducted by heating after specific procedures, starting with the disposal of water and salts and passing through the atmospheric distillation tower and then the vacuum distillation tower to break it down and get different oil products that vary in carbon numbers [1]. After that, it goes through other processes to improve the specifications of the products or to produce other byproducts to make the main product ready for use. Therefore, oil refineries consume a large amount of fuel in refining processes [2]. During each of the stages mentioned earlier (which are subjected to specific circumstances of temperature and pressure) harmful gas emissions, certain oxides, or solid carbon accumulations (particles) are produced. These

emissions or particles spread in many places, such as equipment, furnaces, walls, pipes, heater exchanges, boilers, etc. The combination of these particles reduces the equipment's age and efficiency depending on several factors, most notably crude oil origin, equipment efficiency, refinery capacity, temperature, and refining process [3]. In general, these particles shall be removed because they are harmful and polluting; waste byproducts resulting from the incomplete or partial burning of hydrocarbon compounds in the existing refineries, typically the burnt heavy oil, as well as the ashes resulted, are then land-filled [4]. The development of a novel nanomaterial from available or waste sources with the eligible characteristics [5] has lately grown, and interest in nanotechnology applications has increased [6]. It has a remarkable positive and economic return when using available materials, recycling waste materials, and saving disposal costs by converting them to produce nanomaterials that can be utilized in medical, industrial, and other fields. In addition, these Nanomaterials have unique and exceptional properties, including small size (1 -100 nm) and high surface area [7]. Different techniques have been used to produce Nano material formation, such as the chemical vapour deposition method, hydrothermal synthesis, chemical method, sputtering, spray pyrolysis and others, which rely on the application and the type of materials [8-10]. Among them, sonication crushing has been employed in forming nanostructured material with different sizes related to features such as high-frequency wavelengths. Many research studies have been conducted about extracting carbon nanoparticles or nano-black carbon from various resources or waste materials [11, 12]. In 2013, carbon nanoparticles with sizes of 20-100 nanometers were prepared using the incomplete combustion method of kerosene [2]. Also, a Carbonaceous material has been obtained successfully from spent coffee grounds (SCG), an eco-environmental and economic methodology depended on the mechanical and chemical dry milling, then further carbonization at 800 ºC (it eco-environmental and low-cost approach) in 2018. [13]. J. Alam and others 2021 extracted carbon Nanoparticle from coal fly ash by ultra-sonic, organic solvent extraction, supercritical fluid extraction, soxhlet extraction method, and accelerated solvent extraction because it is a rich source of organic carbons that can be used in various applications [14]. Also, there is much research about applying the used tyres in the production of Nanocarbon, by changing the parameters or the system used in production. For example, in 2019, G. Hernández burned used tyres in a steel reactor at a temperature of 1000 °C to produce Nanocarbon black [15], While in 2021, Chiemeka OnyekaOkoye produced Nano-carbons from waste tyre oil at a very high temperature [16]. This study included a comparison between two samples of petroleum waste (taken from two different places from the Al-Daura refinery) that were considered a high carbon resource and then converted to carbon particles in Nano size by the Sonication method (easy, green, environmentally friendly method) once in the form of a probe and other baths then the Nano carbon particles examined. So, this study presents a novel synthetic approach of nano carbon particles with exceptional properties, especially easy surface functionalization or grafting, and their usage in different applications.

2. Experimental Procedure

2.1 Materials/ Carbon Particle Waste

The petroleum waste samples used in this investigation consist of particles collected from two different locations on Al-Daura refinery equipment during the refining of crude oil and are thought to be a byproduct of the incomplete combustion of hydrocarbon molecules. The atmospheric distillation unit provided the first sample of oil waste, which is used as feed for furnaces that make base oil. During this process, the oil waste goes through various pipes and accumulates at about 270°C - 380°C. Hence, the surfaces of the furnace tubes are used for the first sample (the outer crust of the furnace tubes). The second sample was taken from the residues of the extraction process that involved heating the extract products in a furnace of the furfural treatment unit situated in the strainer refinery before the pump that pushed the material stated for the evaporation unit. When the extract product (an unusual oil product) is heated to about 240 °C, the aromatized compounds with poor lubricating characteristics are removed using furfural as a solvent.

2.2 Methods

2.2.1 Preparation of Carbon Nanoparticles First Sample

The first sample was made by sieving to separate the particles, and the particles were taken that were less than 0.90 mm to obtain a uniform particle size. The powder was cleaned with alcohol and then distilled water to remove impurities such as dust, and it was dried for an hour at 80 °C to finish the purification. Then, it was prepared for EDX-SEM testing, as shown in Fig. 1a. The next step of carbon nanoparticle preparation was reducing the particle size by manually grinding process for about 30 minutes (physical process), which was followed by a chemical process to remove impurities such as some un- required metals, increase carbon content, and decrease particle size to nanoscales, respectively. The process mainly involves burning for an hour at a temperature of 1050 degrees Celsius without oxygen by using thermal pellets coated with ready-made temperature-resistant plaster paste in a vacuum and inert atmosphere, as shown in Fig. 1b. The test is conducted one more time to verify that the percentages are high [17].

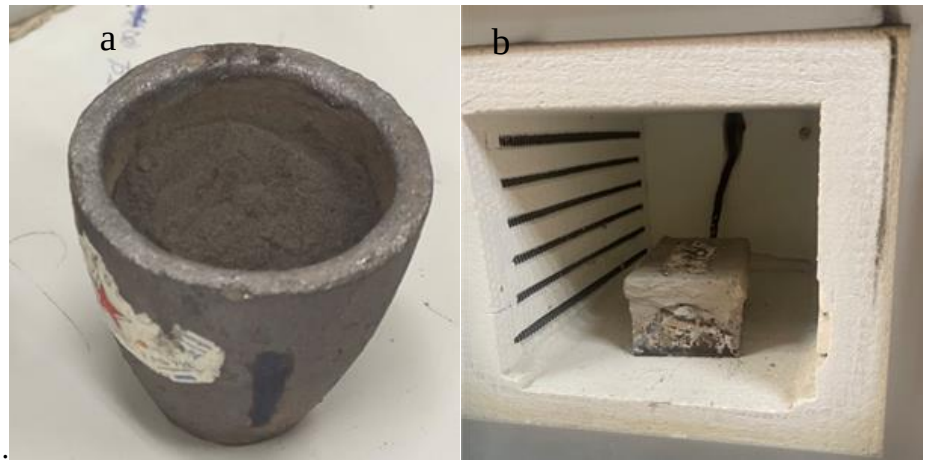


Figure 1: First sample (a), first sample after covering (b).

2.2.2 Preparation of Carbon Nanoparticles Second Sample

The second sample was created utilizing a solvent-solvent procedure to remove the furfural and contaminated material that was present. To ensure that all of the furfural in the sample was removed, it was washed three times with a certain amount of cyclohexane, then was exposed to the atmosphere for 36 hours at room temperature, and dried for the last time in a drying oven at 69 °C for completely drying then sieve less than 0.45 mm to obtain a uniform particle size and the following steps are the same ones used in the sample mentioned above A. The second sample is shown in Fig. 2 before and after being washed with hexane.

Figure 2: The second sample was taken before washing it with hexane.

2.2.3 Preparation of the Samples for Breaking Process by Direct Ultrasonic Process

In this step, four samples—C1 and C2 from the first sample, as well as C3 and C4 from the second sample—are prepared. 2 g of each sample was added to [10 M NaOH] that prepared by (20 g) sodium hydroxide was dissolved in distilled water (50 mL) then stirred until it was homogenous). The C1 and C3 samples were immersed in a 300-watt ultrasonic bath (model: luc-410, Daihan Lab Tech Co., Korea) for 24 hours [18]. C2 and C4 samples were placed in a 900-watt ultrasound probe (model: c202205484, medsinglong global group, china) for 4 hours [19]. The samples are then centrifuged repeatedly and washed with distilled water to remove the hydroxide until the pH is neutralized, the resulting powder is dried, and the samples are ready for testing. Fig. 3a and b shows the preparation of samples by two previous methods.

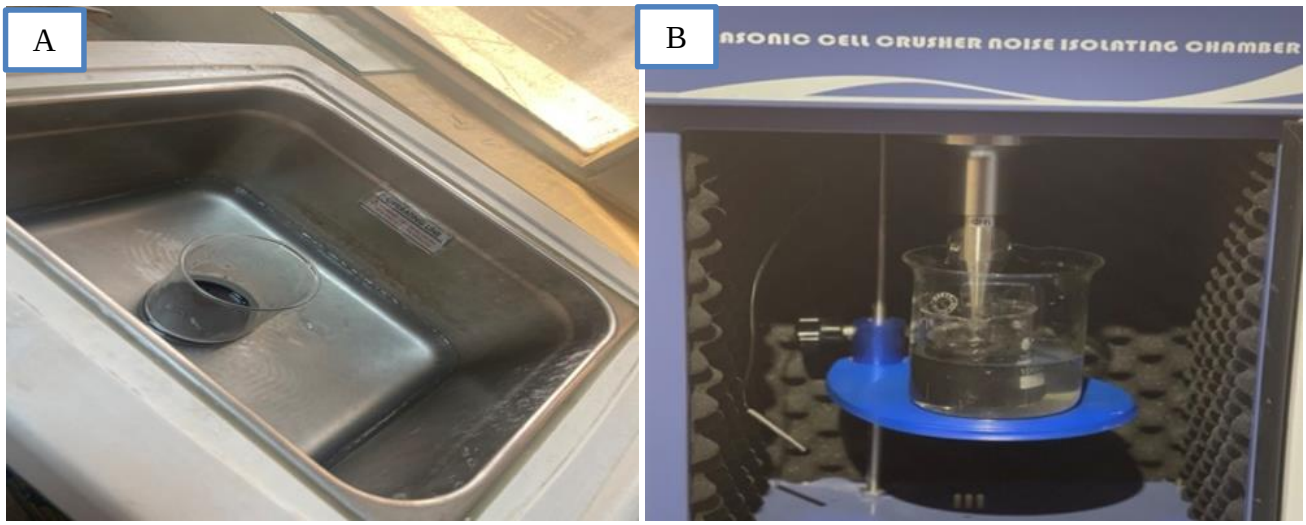


Figure 4: Ultrasound tools: A) ultrasound machine bath samples (C1 & C3), B) ultrasound machine probe samples (C2 & C4).

3. Results and Discussion

3.1 EDX-SEM Exam

Fig. 5 a and b shows the components of the two samples as determined by an EDX-SEM test, and Fig. 6a and b shows the results of an SEM test conducted on samples that had been burned and reduced to nano size (at a temperature about 1050 ^{o}C. As illustrated in Fig. 5, the samples contain significant amounts of unfavourable impurities such as Silicon, Aluminum, Sulphur, and iron. Because the melting point and separation from the raw material are lower than 1000 ^{o}C, most of the impurities, including Aluminum, iron, and sulphur, were released from the samples due to their volatilization. The amount of carbon in the first and second samples, respectively, has dramatically increased from 28.49, & 36.30 to 91.59 & 94.47 wt.% of the total weight, respectively as a result of removal impurities. As a result of the product being submerged in sodium hydroxide during the ultrasonic cracking process, we noticed that a minor amount of sodium and oxygen was identified in the completed product; all the components of the two samples before and after burning are listed in Table 1.

Table 1: Components of the samples A & B before and after burning and after converting to nano size.

Element		Carbon	Oxygen	Silicon	Sulfur	Aluminum	Lead	Sodium	Scandium	Molybdenum
Before burning	A	28.49	36.69	6.30	3.18	1.54	12.42	0	7.61	3.42
After burning		91.59	6.23	0.0	0.0	0.0	0.0	0.89	0.0	0.0
Before burning	B	36.30	37.61	13.53	5.55	4.29	2.71	0.0	0.0	0.0
After burning		94.47	4.42	0.64	0.0	0.0	0.0	0.47	0.0	0.0

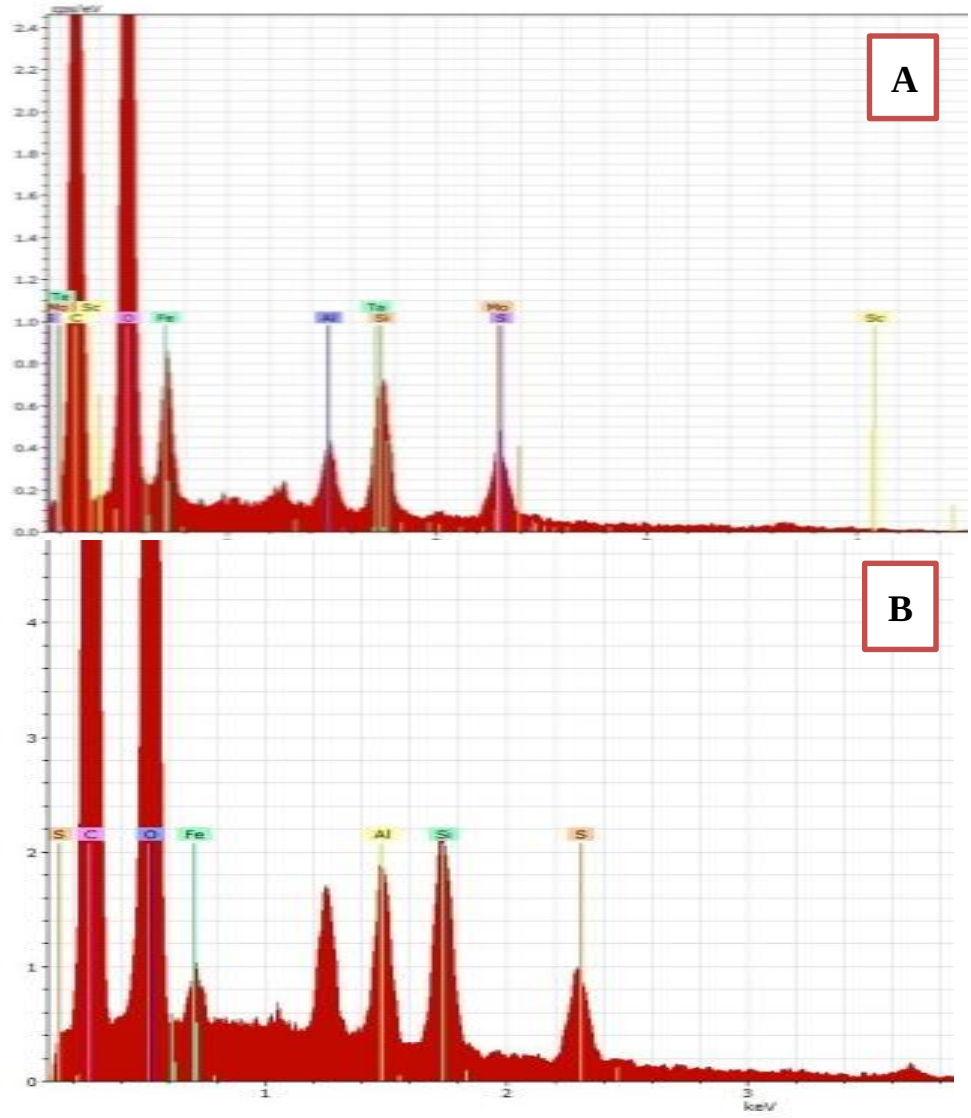
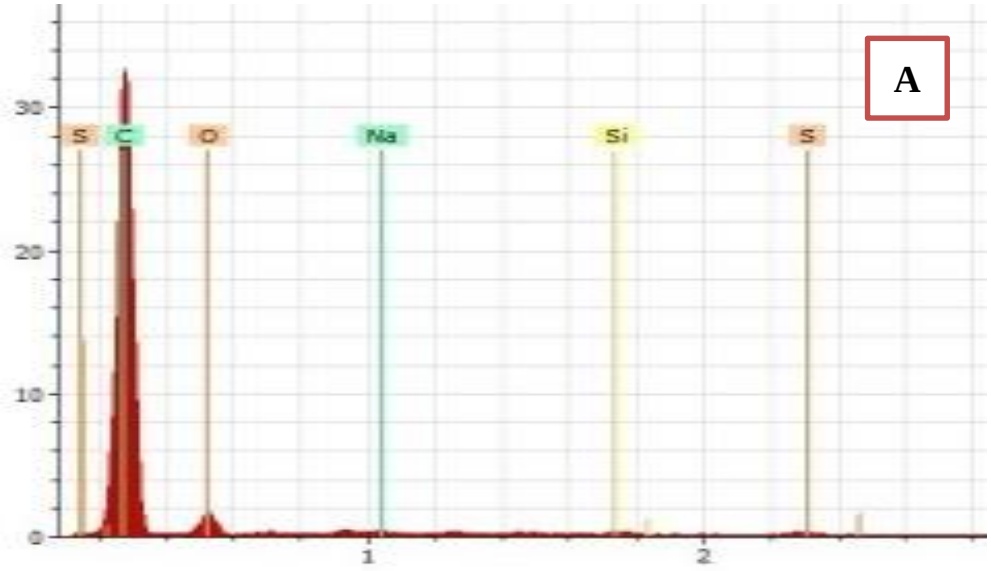


Figure 5: EDX-analysis before burning: A) First sample, B) Second sample.



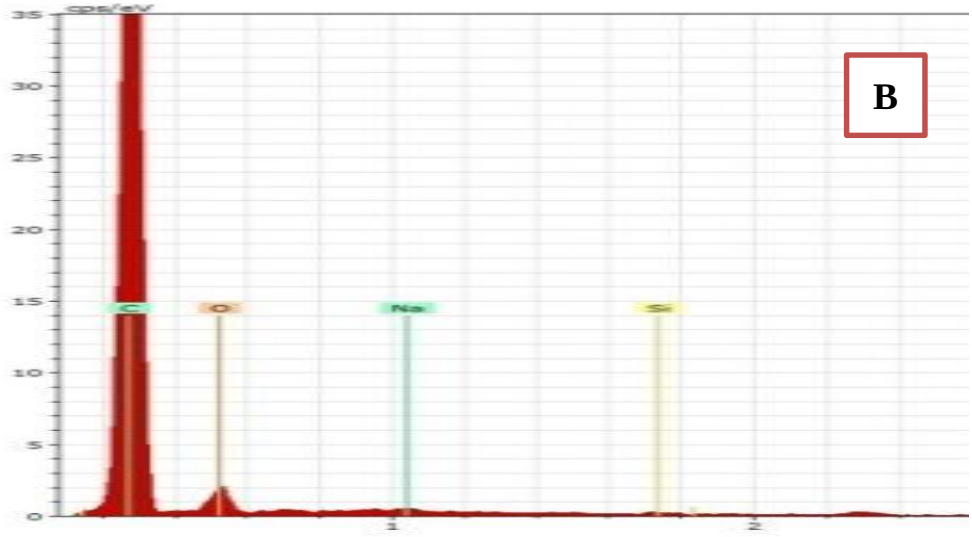


Figure 6: EDX-analysis after burning, Nano size: A) First sample, B) Second sample.

3.2 Particle Size Analysis

Fig. 7 shows particle size analyses for the first, C1 and second samples, C3. The samples' particle sizes decreased from 0.90mm and 0.45mm to 660.8 nm and 223.3 nm. It has been proven that sonication bath treatment (an in-direct ultrasonic procedure) can significantly reduce size but takes longer to achieve Nano size. The nano size was effectively achieved with less time and more power when crushing with a sonication probe (direct ultrasonic process), as illustrated in Figure 8. This is because the direct ultrasonic process has higher efficiency and power than the in-direct process due to the connection between the probe and the particles. On the other hand, for samples C2 and C4, the particle sizes decreased from 0.90 mm and 0.45 mm to 86.8 nm and 37 nm, respectively [19].

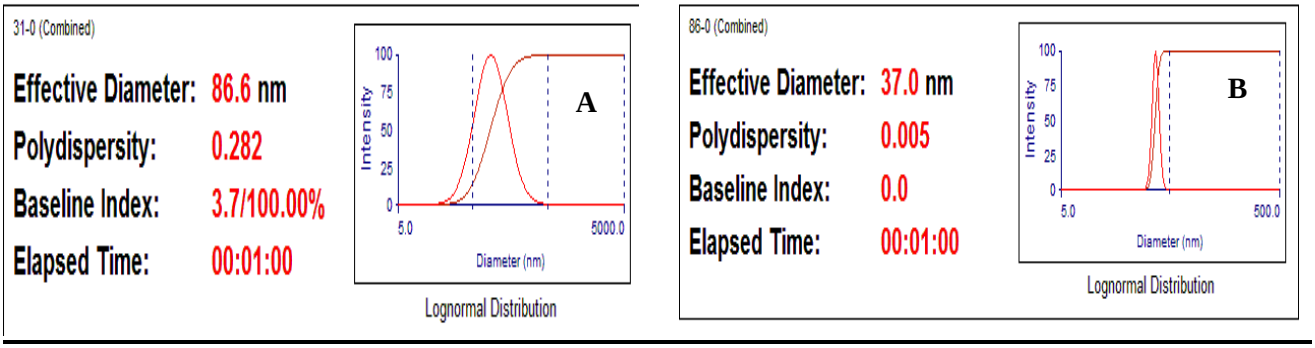


Figure 7: Partical size analyses for sonication bath samples: A) The first sample/ C1, B) The second sample/ C3.

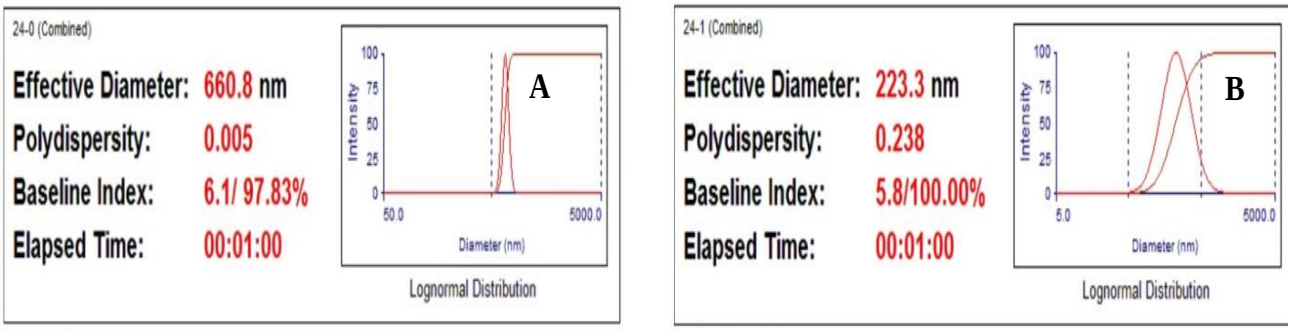


Figure 8: Particle size analyses for sonication prebe samples: A) The first sample/ C2, B) The second sample/ C4.

3.3 SEM Results

Fig. 9 shows the SEM image of unburned samples that, for the first and second samples, respectively, included 28.49 and 36.30 weight present of carbon that were tested after sieving to eliminate all particles larger than (0.90mm) and (0.45 mm). Moreover, the particles' irregularity, their range in size, and the presence of contaminants are noted.

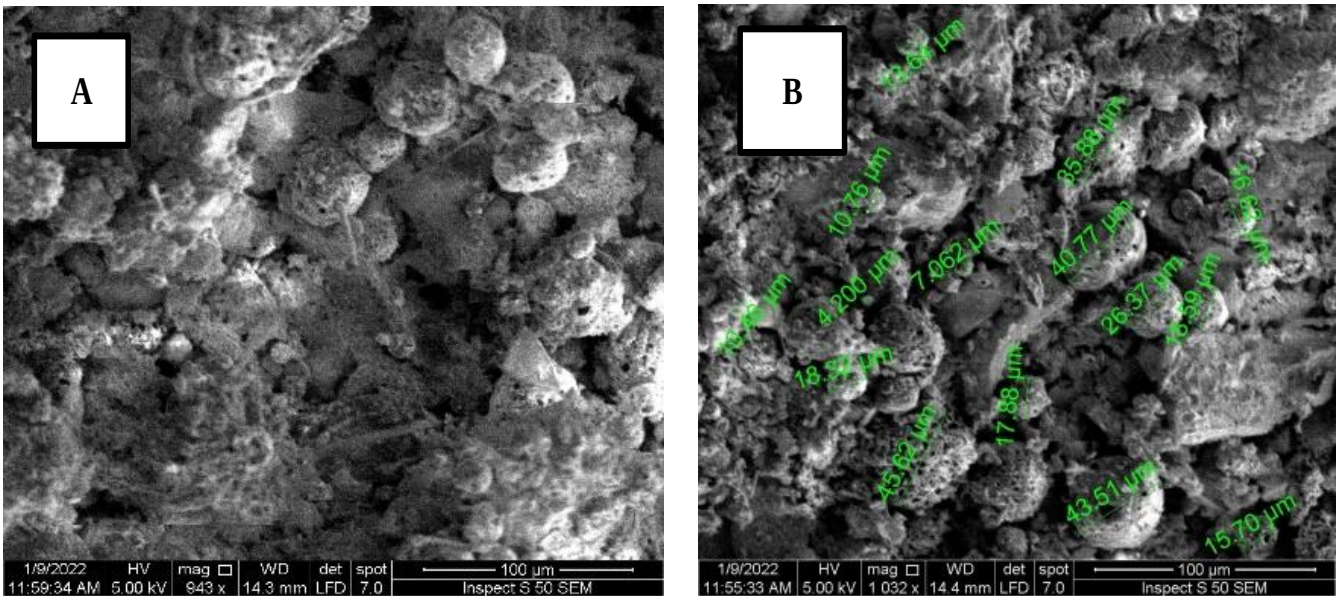


Figure 9: SEM picture of unburned samples: A) First sample / C1, B) Second sample/ C3.

Following the first ultrasonic wave by probe disintegration process and subsequent dispersion operation, Fig. 10 shows the SEM pictures of carbon nanoparticles. The photographs confirmed that crushing using a sonication probe in a base media caused the particles to become nano-sized and exhibited impurity-free particles [20, 21].

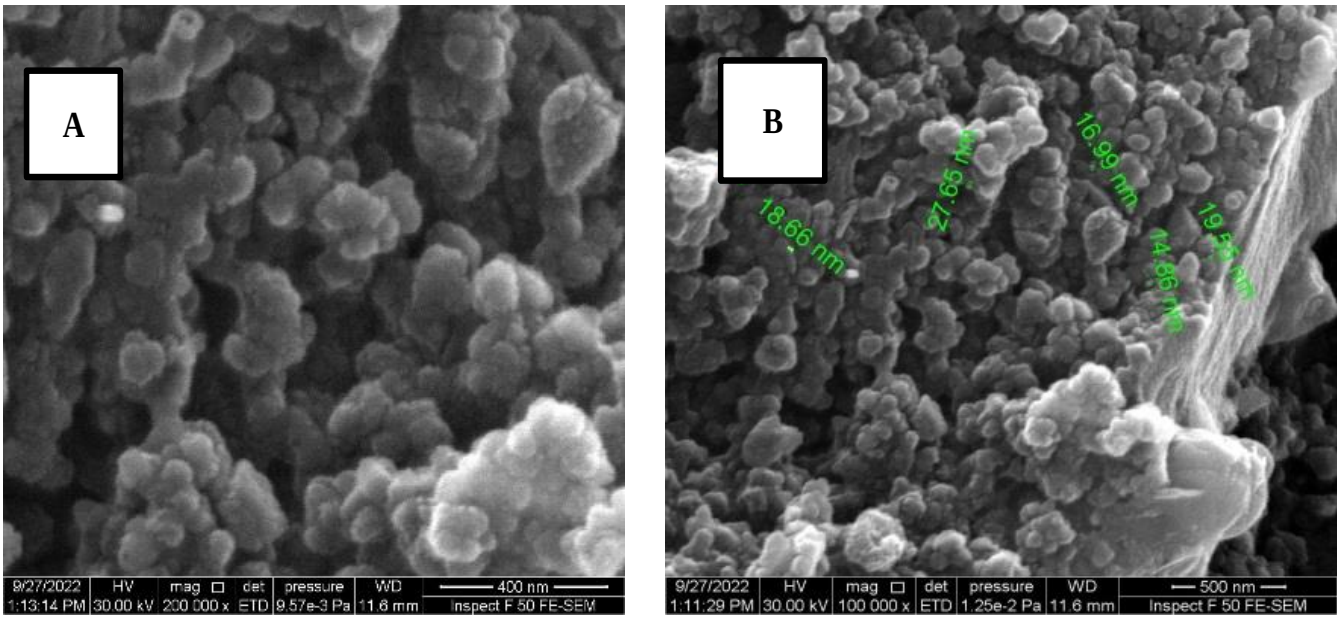


Figure 10: SEM picture of nanoparticles: A) First sample / C2, B) Second sample/ C4.

3.4 Zeta Potential

The dispersion value is around 0.282 (Fig. 11), which is very close to zero and shows that the resulting particles are quite stable, according to zeta potential data, by carefully deciding on the dispersing variables, such as the temperature, the dispersing period, and the particle concentration in the solution.

In addition to the kind of liquid medium used in the dispersing process, excellent stability of the particles formed by the direct cracking process by ultrasonic waves can be attained [22].

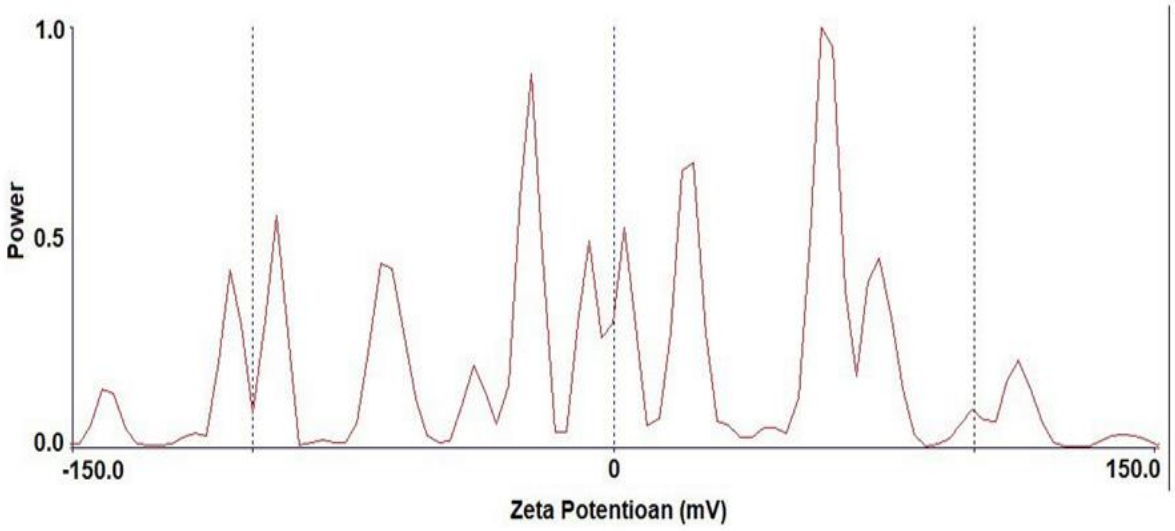


Figure 11: Zeta potential of prepared carbon nanoparticles.

4. Conclusions

Alternatives to chemical or physical treatments in this study: we can turn a polluting and dangerous byproduct into carbon nanoparticles with numerous industrial applications. The crushing by (direct ultrasonic cracking) sonication probe and the indirect ultrasonic process by ultrasonic bath are both practical, inexpensive, and environmentally friendly methods for creating Nanomaterials. Results indicated that the direct ultrasonic approach is more effective than the indirect process at producing carbon nanoparticles. The nanoparticles were created using a direct ultrasonic method, which took less time and made more homogeneous results. Using the ultrasonic dispersion technique in base media, we created nanoparticles with a diameter of less than 100 nm.

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Conflict of Interest

The authors declare that they have no conflict of interest.

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