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Decreasing the nitric oxide gas concentration by using sunflower seed husk biochar as filter

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Abstract. There is a great global interest in controlling air pollutants, specifically greenhouse gases including nitrogen oxides (NO_x), nitric oxide is one of its basic components. For this reason, this study dealt with the use of biochar manufactured from agricultural waste from sunflower seed husk (SSHB) with 450°C pyrolysis in the adsorption of 90 ppm concentration of nitric oxide (NO) gas within 480 seconds (8 minutes) by flow rate 1.2 (liter/minute). Two kinds of particle sizes coarse (C) and fine (F) of SSHB were used, and some physiochemical properties of the biochar were studied, including SEM and FTIR, in addition to the BET surface area, which was 3.9 (m^2/g). The adsorption results displayed that SSHB (F) is better at the adsorption of NO 31.6 (mg/g) than SSHB (C) 26.7 (mg/g). Isotherm models were applied to the mathematical modelling of NO absorption, and based on n and R^2 values the results of the SSHB adsorption for NO fixed with both the Freundlich model and the kinetic pseudo second order model.

Keywords: Air Pollution, Sunflower Seed husk, Biochar, Nitric Oxide, Adsorption Isotherm.

1. Introduction

One of the greatest significant threats to environmental quality worldwide is the tricky of air pollution with its undesirable belongings on the environment and humans. Air pollution can be defined as any lengthy attendance of great concentrations of pollutants. In general, any chemical, physical, or biological compound that negatively affects the quality of the atmosphere is considered an air pollutant [1]. NO_x, CO, SO_2, and particulate are considered primary air pollutants. While ozone, carbon dioxide, and sulfur trioxide are considered secondary air pollutants. Nitrogen oxides (NO_x) are among the most studied compounds in air pollutants, as they are one of the emissions from many sources of pollution. They are considered colorless, odorless, toxic gases that are not soluble in water. They remain for several days when emitted into the atmosphere, and ninety percent of them are nitric oxide (NO) [2]. Therefore, it was necessary to think about developing strategies and solutions to reduce and control air pollutants. Adsorption technology is the most widely used treatment technology used to reduce pollutant concentrations. For example, there are common materials that are used as adsorbents and have been used in the adsorption of inorganic and volatile organic pollutants, including active carbon and zeolite, in addition to other types of materials such as polymers [3]. Some studies have shown the ability of activated carbon to adsorb VOCs with a removal rate of up to 90% for BTEX [4]. Carbon nanocomposites also showed a removal capacity of up to 140 μg/m^2.h [5]. By catalytic oxidation, some carbon materials, such as activated carbon, have been used to reduce nitrogen oxides [6]. Due to the abundance of agricultural waste and the low price, there is constant research into developing the production of effective, sustainable, low-cost carbon absorbent materials (biochar) [7]. Biochar is a solid plant material that is converted into carbon by pyrolysis of biomass in the presence of little or no oxygen. Recently, biochar has been used in comparison with activated carbon in adsorption techniques in gaseous environments [8]. Sunflower seed husks are agricultural waste that is a byproduct of oil production and can be used to convert them into biochar. Biochar from sunflower seed husks has been used in several processing techniques as an adsorbent in aquatic environments to remove the elimination of pollutants [9]. In the current study, biochar was manufactured from sunflower seed hulls without activation using advanced techniques to test its efficiency in the adsorption of nitric oxide gas, an important gaseous pollutant and, to study its ability to compete with other biochar raw materials and commercial materials with high manufacturing costs in using as adsorption material that is available and inexpensive to produce.

2. Materials and Methods

2.1. Biochar preparation as adsorbent material

To manufacture biochar was collected sunflower seed husk (SSH) residue, then washed with tap water, then deionized water to remove unwanted materials and, air-dried. By pyrolysis in a muffle VULCANE A-550 furnace biochar was manufactured from sunflower seed husk (SSHB) for two hours at a temperature of 450 ± 5 °C in a limited environment of O_2. After cooling the SSHB were ground with stainless steel sieve into coarse particles (C) of size (1- 0.5) mm and fine particles (F) of size (< 0.250) mm as shown in Figure 1.

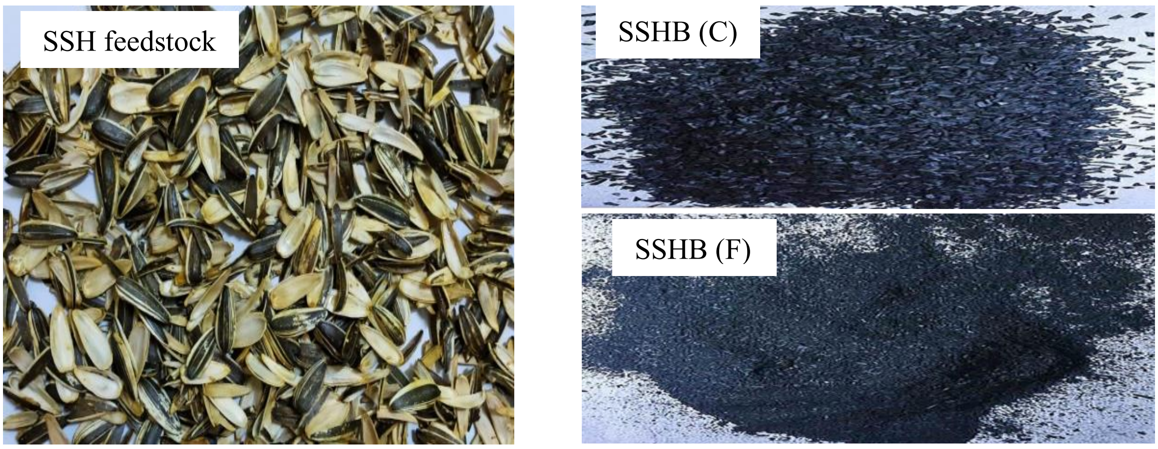

Figure 1. SSH before and SSHB after pyrolysis process to produce biochar and grinding to two particle sizes C and F.

2.2. Characterization of sunflower seed husk biochar (SSHB)

To study the physicochemical properties of SSHB, the yield value, ash content and, moisture content of SSHB were estimated [10]. The Beckman Coulter SA(TM) 3100 analyzer was used to estimate the surface area and pore volume of SSHB(C) using the Brunauer Emmett Tiller (BET) law to estimate the surface area, and the Barrett Joyner Hallender for pores size and its distribution [11]. The surface morphology of SSHB (C) was also studied using a scanning electron microscope (SEM) type (Phillips SEM-505) for the accelerating voltage of the device 20 kV. The structural properties and functional groups of SSHB (C) were also determined using an FTIR spectrometer (FT/IR-5300) in the range 400–4000 cm^{-1}.

2.3. Filtration System (FS)

A canister plastic cylinder with 468.7 cm^3 of volume and dimensions of length (19.7 cm) and, diameter (5.8 cm) was used. Contains two barriers (filter paper and metal filter) and is controlled by the canister volume install the barriers by spiral metal, with a space (free of adsorb eat materials) on one side of the cylinder for the gas to enter and exit.

2.4. Adsorption experiment

The concentration of a standard cylinder of NO gas was estimated by connecting it directly to the Gasmet gas analyser FTIR device at a flow rate of 1.2 (liter/minute) to ensure its concentration (90 parts per million), and the reading was estimated as a blank value for the samples. The FS canister was then filled one time with a biochar sample with a fine particle size SSHB (F), and another time with a biochar sample with a coarse particle size SSHB (C) and connected each time to both the NO gas cylinder and gas analyser device, and a gradual increase in concentrations until reaching a steady state was observed. The concentration readings were recorded over 480 seconds (8 minutes). This process was repeated more than once with new samples of each type to ensure the accuracy of the readings. Figure 2 shows the design of the measurement experiment as well as the gas flow path from the cylinder to FS to the unit of measuring gas concentration.

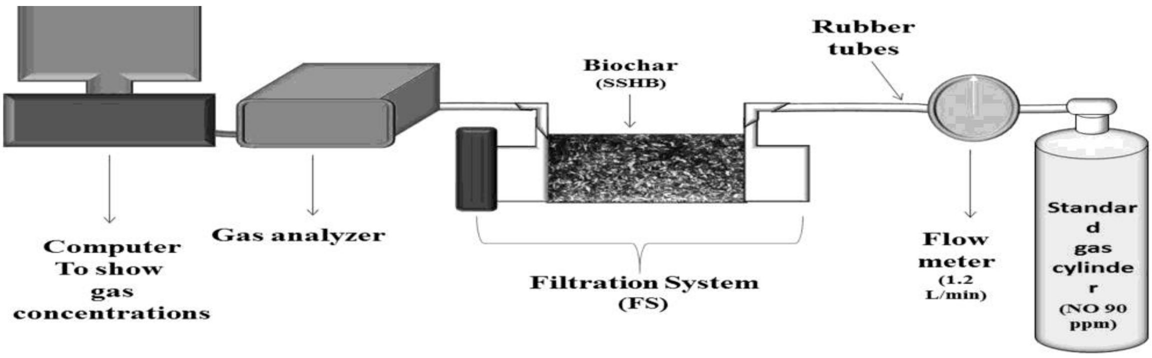

Figure 2. Filtration system diagram.

2.5. Analysis of adsorption experiment data using isotherm and kinetic models

To analyze the results were represented on isotherm models. The Langmuir model explains the nature of adsorption on an active and stable monolayer area where there is a homogeneous surface, as it retains one molecule without interacting with the rest of the other molecules at a time [12]. The Freundlich model which assumes that adsorption occurs on a heterogeneous surface with an irregular distribution of heat of adsorption [13]. The Temkin model shows that due to adsorbent interactions with surface saturation, the temperature of adsorption in the layer decreases linearly for all molecules [14]. To know the mechanism of adsorption on adsorbed surfaces, kinetic models are applied to experimental data to determine the appropriate kinetic model. The best fitting kinetic model is determined based on r^2 values that lie between 0-1, where the value closest to one is the best fit. It used a pseudo first order model according to Lagergren, a pseudo second order model [15].

3. Results and discussion

3.1. Characterization of SSHB

As shown in Table 1, some physicochemical properties of sunflower seed husk biochar (SSHB) that was produced at 450°C for two hours. The ash content was 5.7% and the moisture content was 4.8%, while the biochar yield was 39.5% and the percentage of volatile gases was 50%. The estimating the Brunauer Emmett Teller (BET) surface area, total pore volume, and pore size distribution of SSHB, specifically the coarse volume C, the results showed that the pyrolysis of SSH at 450°C contributed to the presence of a good porosity in the manufacture of biochar, as the surface area was 3.9 m^2/g, and volume all pores was 12.5 mm^3/g. This is what was shown by examining the compositional interface and morphology of the SSHB C surface by SEM at 1 μ magnification as shown in Figure 4. As is clear from the results, the size and shape of biochar particles have an impact on their ability for adsorption, and this is what some previous studies have shown when using other types of biochar with different particle sizes when adsorbing both methylene blue and iodine that is according to the study of Mohamad et al., which the adsorption capacity of biochar towards small particles has been proven and this is attributed to the increase in the adsorption surface area with small particle size [16]. The results for the functional groups of SSHB are shown in Table 2 and Figure 3, the FTIR spectra had bands at 830 cm^{-1} where the alkenes are C-H, 1391.2 cm^{-1} where the asymmetric C-H and aromatic C-C are, 1581.2 cm^{-1} where the amine group is, 2350.8 cm^{-1} where the extended hydroxide is, and 3419.2 cm^{-1} where the kohl and phenol groups are. The raw material for SSHB consists of lignocellulose polymers, which are divided into lignin, cellulose, and hemicellulose. These components contain groups of (carboxyl, hydroxyl, carbonyl, amine, and other groups) that are surface active, which helps in the presence of a surface area for the reaction, which has a role in the adsorption process, and this is according to what was shown in the study of Soldatkina et al. (2009) Spectra of SSH [17].

Table 1. Physicochemical Parameters of SSHB.

	SSHB F	SSHB C
Surface area (m^2/g)	-	3.9
Total pore volume (mm^3/g)	-	12.5
Volume of mesoporous and macro-porous (mm^3/g)	-	9.8
Volume of microporous (mm^3/g)	-	4
Biochar yield %	39.5	
Ash %	5.7	
Moisture %	4.8	
Volatile gases %	50	

Table 2. FTIR wavenumbers of SSHB.

Groups	Number of waves (cm^{-1})
C-H bend alkenes	830.3
C-H asymmetric deformation & C-C aromatic	1391.5
N-H bend	1581.5
O-H bend	2351
O-H alcoholic and phenol, NH	3419.5

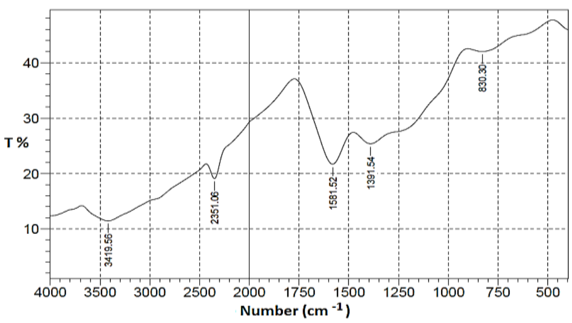

Figure 3. FTIR peaks wavenumbers of SSHB.

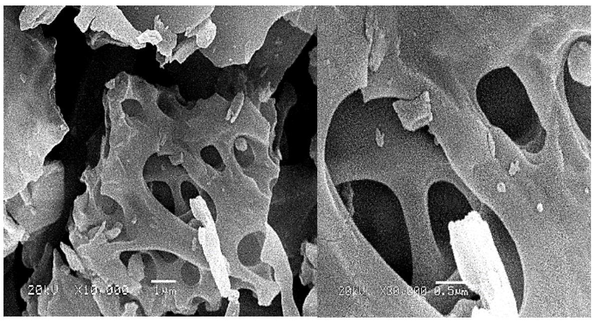

Figure 4. SSHB (C) scanning electron microscopy image with the scale of 1μ magnifications.

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3.2. Nitrogen oxide (NO) absorption on sunflower seed husk biochar (SSHB)

Results for NO concentrations after adsorption on SSHB were obtained in both fine (F) and coarse (C) particle sizes, as shown in Table 3 and Figure 5, whereas the results showed that the ability of SSHB F to absorb NO gas at a concentration of 90 ppm is 31.58 (mg/g) within 480 second (8 minutes), while the ability of SSHB C to adsorb NO gas at a concentration of 90 ppm was 26.68 (mg/g) within 480 second (8 minutes). These results, it was shown that the ability of SSHB F is higher than the ability of SSHB C to adsorb NO gas. Also, evident that the adsorption process is affected by the size shape of SSHB particles, due to the difference in the kinetic diameters of the gases N_2 (0.364 nm), NO_2 (0.34 nm), NO (0.317 nm), and their steric effects on filling the pores upon adsorption, this may have a clear effect on the efficiency of adsorption with the difference in pore size. It has also been proven that there is an effect related to the Vander Waals constant on the extent to which the size of the micropores is filled so that in the size of the micropores, the chance of adsorption of both N_2 and NO is much less than the chance of adsorption of NO_2 [18].

Table 3. Adsorption capacity of SSHB with two particle sizes F and C for NO gas (Q mg/g) and NO concentration after adsorption (Con ppm) and in time (second).

Time	Blank Con	SSHB			
		F		C	
		NO Con	SSHB Q	NO Con	SSHB Q
30	74.3	1.26	0.47	9.25	0.57
60	83.5	18.4	0.89	35.74	0.98
90	83.5	39.9	1.17	55.21	1.23
120	84.5	56.1	1.36	66.74	1.39
150	86.2	55.2	1.56	69.13	1.53
180	87.8	60.7	1.73	73.3	1.66
210	85.6	59.8	1.90	77.84	1.73
240	88.9	70.1	2.02	81.2	1.80
270	88.7	62.9	2.18	84.37	1.83
300	87.4	72.5	2.28	80.12	1.90
330	86.2	76.7	2.34	82.62	1.93
360	89.6	65.3	2.50	82.84	1.99
390	85.8	63.7	2.64	84.94	1.99
420	87.3	65.09	2.78	84.98	2.01
450	89.5	76.2	2.87	83.71	2.06
480	83.9	80.9	2.89	82.09	2.08
Total Q in 480 second			**31.6**		**26.7**

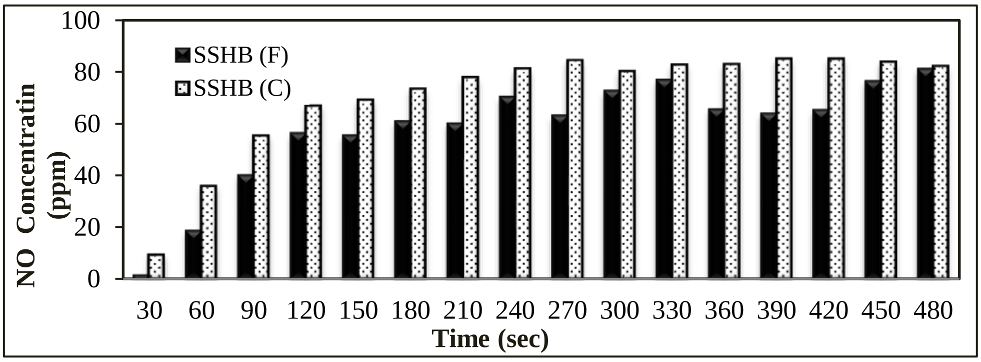

Figure 5. NO gas concentrations during its adsorption on SSHB with two particles sizes (F, C).

As shown in Table 4, a comparison of the adsorption capacity of SSHB for nitric oxide gas with the same capacity for other species under similar and different experimental conditions. SSHB has shown good adsorption capacity compared to some types, for example, Rice husk biochar 500ºC (F). It also gives a lower ability to adsorption with some other species. This means that many factors affect gas adsorption, including the type of production material, and the size of its particles. There is also an effect due to the vacuum shape of the filtration system that is used in the measurement. However, considering the traditional method of biochar production from sunflower seed husk (SSHB) that was used in this study, this indicates the possibility of increasing the adsorption efficiency of SSHB when using methods of chemical or physical activation for it, which contributes to increasing the adsorption capacity.

Table 4. Comparison of SSHB in the current study with various types of other carbon adsorbents in previous studies in its ability to NO gas adsorption.

Adsorbent	NO concentration (ppm)	Adsorption time	Flow rat. (l/minute)	Adsorption capacity (mg/g)	References
SSHB 450 ºC (F)	90	480 second	1.2	31.6	Current work
SSHB 450 ºC (C)				26.7	
Rice husk Biochar 450 ºC (F)	90	480 second	1.2	49.5	[16]
Rice husk Biochar 450 ºC (C)				48.2	
Rice husk Biochar 500 ºC (F)				25.1	
Rice husk Biochar 500 ºC (C)				40.5	
Activated carbon by Cerium for palm shell	500	-	-	3.5	[19]

F: fine particle sizes, C: coarse particle sizes

3.3. Adsorption isotherm studies

The adsorption mechanism of NO gas on SSHB was studied by characterizing the adsorption isotherm data for Langmuir, Freundlich, and Temkin models. As is evident in the isotherm curve in Figure 6 and as is evident from the parameters of the isotherm models in Table 5. The Freundlich model showed a better fit to the adsorption data as the n values were 2.3 and 1.7 for SSHB(F) and SSHB(C), respectively. The values of n show good agreement at 2-10, average at 1-2, and weak agreement at < 1, according to Tribal [20]. While the R^2 values were low compared to the other models, which means that they did not fit well. In general, agreement with the Freundlich model means that adsorption occurs through physical and chemical processes on a heterogeneous, multilayer surface of the adsorbent [21].

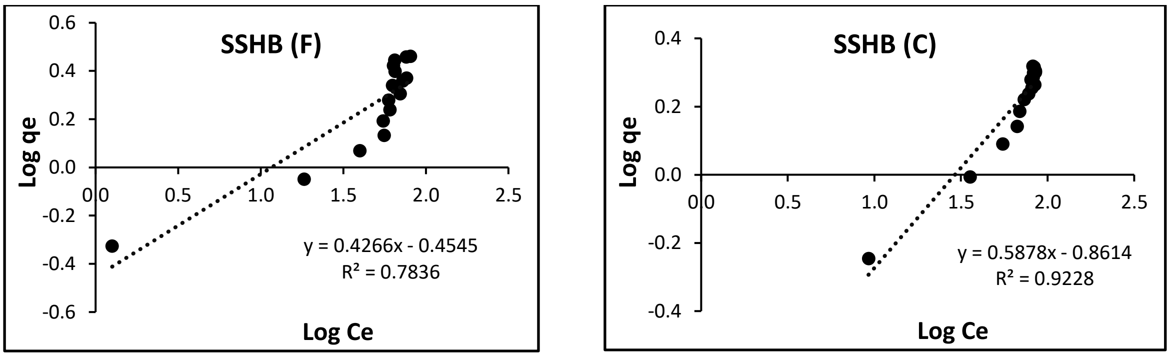

Figure 6. Freundlich isotherm curve for SSHB adsorption of NO.

Table 5. Isotherm models for SSHB adsorption of NO.

SSHB samples		Constants of Langmuir			Constants of Freundlich			Constants of Temkin		
		K_L	q_m (mg/g)	R^2	K_F	n	R^2	B	A	R^2
SSHB	F	0.02	4.0	0.4	0.4	2.3	0.78	0.5	0.9	0.5
	C	0.01	3.7	0.3	0.1	1.7	0.92	0.7	0.2	0.79

3.4. Studies of kinetic adsorption

As shown in Table 6, three models were used to determine the adsorption dynamics as well as estimate the residence time in the adsorption process. As shown in Table 6, the pseudo first order model constants k^1 and q_e are determined by the intersection q_e-q_t with t, the constants of the pseudo second order model K^2 and q_e were determined by the intersection of t/q_t with t. It was clear from the values shown that the most consistent model for the results of the experiment is the pseudo second order model, where R^2 represented its highest values, which were 0.986 and 0.999 for both SSHB (F) and SSHB (C), respectively as shown in Figure 7. In general, agreement with the pseudo second order model indicates the possibility of a chemical nature in adsorption [22].

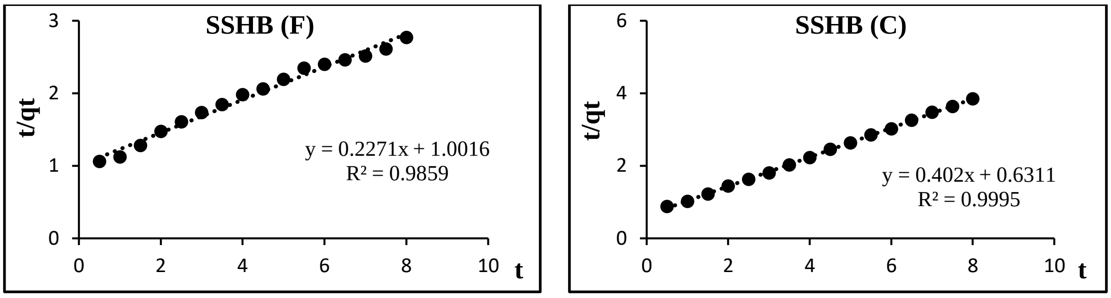

Figure 7. Linear regression of pseudo second order kinetic model for SSHB adsorption of NO with two particle sizes fine (F) and coarse (C).

Table 6. Kinetic models for SSHB adsorption of NO.

Biochar Sample		Simple first order			Pseudo first order			Pseudo second order		
		K_1	C_0	r^2	K_1	q_e	r^2	k_2	q_e	r^2
SSHB	F	0.3	14.9	0.38	-0.7	8.5	0.66	0.1	4.4	0.986
	C	0.2	33.4	0.43	-0.5	2.1	0.94	0.3	2.5	0.999

4. Conclusions

The physicochemical properties of sunflower seed husk biochar (SSHB) showed its ability to adsorb, which is due to the porosity of the surface area. This was shown by estimating the surface area, as well as SEM images and FTIR functional groups to study the surface of SSHB. When different sizes of SSHB particles were used in the experiment, it was found that they affected the quantity of adsorption, whereas particle size fine (F) was better than the coarse (C) in absorbing nitric oxide gas (NO) on SSHB. When describing the results with isothermal models, it was found that the most suitable model to designate the NO adsorption process on the SSHB surface is Freundlich. When studying the kinetic models, it became clear that the pseudo second order best describes the adsorption kinetics on the SSHB (C) and SSHB (F) in this experiment. Overall, SSHB prepared by traditional methods showed a good response in the adsorption of NO gas under laboratory test conditions, which means that there is an ability to use SSHB as an adsorbent for NO gas. It can be recommended to study the possibility of increasing the adsorption efficiency of SSHB by using advanced manufacturing techniques, such as the activation of biochar. It can also be recommended to conduct further studies on biochar from agricultural wastes to use them as an available, inexpensive, and effective alternative in controlling air pollutants gas in general and nitrogen oxide gas in particular.

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