

Research Article

Adsorption of SO₂ on ZnO Nanowires Using Activated Carbon by Langmuir Adsorption Isotherm

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Sulphur dioxide (SO₂) is one of the contaminants present in the environment. They are the by-product of combustion, industrial pollution, generation of electricity, etc. After some reactions in the atmosphere, SO₂ changes its form and produces acid rain. The presence of SO₂ needs to be detected to combat its effect. Many nanosensors are designed to detect the presence of SO₂ in the environment. Zinc oxide (ZnO) nanowire sensor is one of those sensors used for SO₂ detection. The size, structure, cost-effectiveness, and unique properties made it a choice for sensing purposes. Activated carbon is another compound that has porous substances that helps the adsorbate to settle down on the large surface area of its adsorbent surfaces. Combining the nature of ZnO nanowire and activated carbon, the adsorption of SO₂ can be increased. This paper proposes a novel technique involving the activated carbon in the ZnO nanowire sensor to increase its SO₂ adsorption capacity and rate. The Langmuir adsorption isotherm is used to find the adsorption efficiency between the solid adsorbent and gaseous adsorbate. MATLAB simulation was carried out for the proposed work in which it is seen that the novel method shows 33.34% efficiency in terms of SO₂ adsorption capacity. The response of the proposed sensor shows 23% efficiency over time. The analysis shows that the usage of the activated carbon increases the adsorbent site for SO₂ adsorbate to adsorb on the surfaces. According to the adsorption quantity, SO₂ level has been obtained in the environment.

1. Introduction

One of the indirect greenhouse gases is sulphur dioxide (SO₂). The SO₂ contributes less percentage in greenhouse gases, but it has hazardous effects on nature. The sulphates are the by-product of SO₂ which leads to acid rain [1]. The derivatives of SO₂ when it reacts with the atmosphere are even worse. The provenance of SO₂ is mainly combustion fuels, refineries, industrial boilers, electricity generation, diesel engines, metal processing, etc. The Eastern States are mainly affected areas due to this SO₂. It causes respiratory

illness and leads to some cardiological effects [2]. To reduce the effect of SO₂ or combat SO₂, the main thing needed is to observe the places where it is present. Wireless sensor networks play a major role in sensing many particulars in unmanned areas.

The nanowire sensors are very tiny with a 1D structure [3]. Many kinds of nanowire sensors like metallic nanowire sensors, metal oxide nanowire sensors, metal nanowire gas sensors, and silicon nanowire gas sensors are evolved. This paper uses a ZnO nanowire gas sensor to detect the presence of SO₂ in the atmosphere [4]. For determining the sulphur

dioxide concentration in the exposure chamber, known volumes of the gas-air mixture were taken periodically with the help of a device known as an aspirator and analyzed. This suction apparatus is connected to a rotameter to regulate the gas-air mixture's suction. The selection of the ZnO nanowire sensor is due to the solid-gas interaction. Usually, the nanowire sensors work efficiently at low temperatures. The characteristics will change when the temperature varies. The ZnO nanowire sensors are widely used for SO₂ detection. The Langmuir adsorption algorithm isotherms explain how pressure affects the gaseous adsorbate on the solid adsorbent [5]. Zhang et al. discuss the various manufacturing techniques of ZnO nanowires, their features, and their applications. The paper mainly focused on the photocatalytic property of ZnO nanowires which helps to protect the environment [6]. The food aroma detection and identification of solvents are discussed by Kohl [7]; the author discussed the temperature dependent of open reactive surfaces of the gas sensors. Cui gave the survey of ZnO nanowires, including their structure, functionality, area of research, and future applications. The p-type doping of ZnO nanowires was also provided [8]. Ramgir et al. presented the survey of recent developments of nanowire-based sensors and discussed different types of nanowire sensors and the efficiency of the array of nanowire sensors for future medicinal purposes [9]. The biological and chemical species are studied using the nanowire sensors which have the property of ultrasensitive and significant way of using the nanowire sensors for diverse applications [10]. Zhang et al. describe the top-down and bottom-up approaches for detecting chemicals and explain various metal oxide sensors and their applications. The QSAR model is used to study the adsorption of SO₂ on the ZnO nanowire sensors explained in paper [11]. Li and Ma elucidate the mechanism of removal of SO₂ using the activated carbon. The temperature-dependent characteristics were also studied [12]. Dunicz explained that the specific area of adsorption of charcoal is studied using Langmuir Adsorption isotherm [13]. Many studies are now available showing the effects of gaseous and particulate pollutants emitted from power plants. However, the effects of those pollutants on human health and animals are less extreme, which does not constitute a direct health hazard. Given the global increase in atmospheric change (CO₂, CO, NO, and acid gases), the consequences of a massive increase in the use of coal, while not fully understood, are likely to be disastrous. Novel of this research work is was aimed at investigating the usage of the activated carbon on the surface of ZnO nanowire sensors to reduce the porous dimensions to enhance the absorption rate of SO₂. The solid-gas interaction-based Langmuir algorithm is used to estimate its performances.

2. Materials and Methods

2.1. ZnO Nanowires. The unidimensional property of nanowires yields interest for many electrical, optical, electrochemical, and mechanical applications. Among the different nanowires, ZnO nanowire has unique properties that show remarkable performance in various applications [14]. The

important feature of ZnO nanowire is its exciton binding energy which is high, and the surface to volume ratio is also large. The quantum confinement effects of ZnO nanowires makes the transport of photons, electrons, and carriers. The ZnO-based nanowires are the richest among the other nanostructures because of their structures and properties [15]. There are two main structures of ZnO nanowires: hexagonal wurtzite and cubic zinc blende. The wurtzite structure of the ZnO nanowire is more stable at ambient temperatures. The ratio of planes approximates the unity that forms the hexagonal structure, making its faster growth in three different directions and helps in adjusting the area of the facets in the same three directions. The ratio $c/a = 1.633$ which equals unity, thus forming HCP structures. The structure of ZnO nanowires is studied using scanning electron microscopy, X-ray diffraction patterns, and transmission electron microscopy. The studies show that ZnO nanowire is a single crystal structure with the $\langle 001 \rangle$ growth direction; it grows vertically on the substrate with 100 nm diameter. The negative field effect only enhances the properties of nanowires, and the diffraction pattern suggests that they are single crystals [16].

There are two methods employed for the synthesis. Figure 1 shows different methods for synthesis. The first method is vapor phase synthesis (VPS). The vapor species are produced in the closed chamber by evaporation, gaseous reduction, and chemical reaction at very high temperatures 500°C to 1500°C. The different types of vapor phase synthesis are vapor liquid solid growth (VLS), chemical vapor deposition (CVD), metal-organic chemical vapor deposition (MOCVD), physical vapor deposition (PVD), molecular beam epitaxy (MBE), pulsed laser deposition (PLD), metal-organic vapor phase epitaxy (MOVPE) [17]. The MOCVD and VLS have widely used techniques for synthesis.

The VLS method is a cheaper and easy process for manufacturing large wafers. Another type is solution phase synthesis, which comprises three techniques: hydrothermal, microemulsion, and ethanol-based. The solution-based synthesis uses organic and inorganic substrates using the aqueous solution or organic solution for the growth processes [18]. It is quite simple, cost-effective, and easy to manipulate. There is also one type of growth called patterned growth, where arrays of nanowires are grown to meet optical properties for lasers. The properties of ZnO nanowires are spontaneous emission, stimulated emission, electrical properties, piezoelectric properties, dilute magnetic properties, etc. The ZnO nanowires are n-type material because of four oxygen connected to two zinc ions interstitially [19]. The FET is designed using ZnO nanowires where the back gate is present. The n-type and p-type shows different responses and characteristics for the same inputs [20]. The applications of ZnO nanowires are gas sensors, biosensors, UV detectors, UV laser, light-emitting diodes, dye-sensitized solar cells, nanogenerator, field emission device, field effect transistors, photocatalysis, photodetectors, ferromagnetism, electricity generator, etc.

2.2. ZnO Nanowire Sensor. There are two approaches by which nanowire sensors are fabricated: top-down and

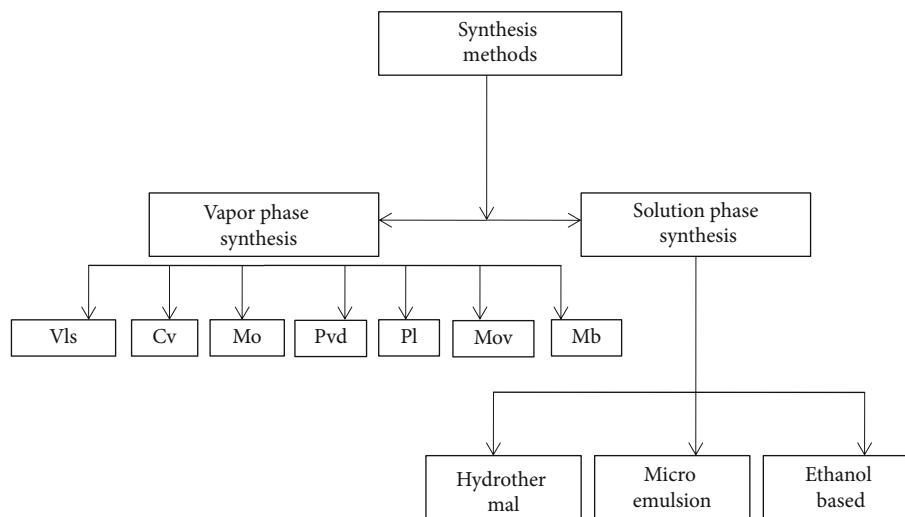


FIGURE 1: Different types of synthesis methods of ZnO nanowires.

bottom-up. In the ZnO nanowire sensor, the silicon substrate is formed on the surface, which adsorbs the chemicals gases present in the environment. The adsorption rate of the ZnO nanowire sensor based on the redox reactions induced by the charge transfer process is made. The selectivity and sensitivity of the ZnO nanowire sensor are very high so that it will particularly adsorb the chosen gases from the environment for which it is designed. The main reason for high sensitivity is gas diffusion at porous and shell layers, which leads to electron depletion [21]. The ZnO nanowire sensor obtained using the thermal vaporization method yields more efficiency and faster recovery. The array of ZnO nanowire sensors helps to detect H_2S in the environment. The ZnO nanowire sensor act as FET with the back gate electrode. The back gate electrode by applied voltage increases the sensitivity of the sensors [22]. The sensitivity of the ZnO nanowire sensor is directly proportional to the electric potential applied to the back gate. The full refreshment of the ZnO nanowire sensor is done by applying -60 V as the back gate voltage. The dimension of ZnO used in a FET varies from 20 to 60 nm. The amount of charge transfer examines the ZnO surfaces and SO_2 interaction. Depending on the charge value and its direction, the function of the sensing material will be described [23]. The highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) define the interaction amount for different concentrations of SO_2 exposed. By studying the Fermi energy level, the energy barrier is reduced by applying the back gate voltage, which helps the sensor adsorb more SO_2 on the surface [24].

2.3. Activated Carbon. Activated carbon is tiny and low-volume porous materials of powdered or granular form. The pyrolysis and chemical treatments are the way to produce activated carbon using coal, coconut shells, bamboo, organic materials, etc. With its low-volume pores, the activated carbon increases the substrate's surface area, which helps the adsorbate interact at the porous site created [25]. The pore diameters of the activated carbon show heteroge-

TABLE 1: Simulation parameters.

Parameters	Merits
Intrinsic carrier concentration	$<10^6\text{ cm}^{-3}$
Effective mass	0.24 (e^-) and 0.59 (holes)
Exciton binding energy	60 meV
Band gap energy	3.3 eV
Dielectric constant	8.66
Lattice constants	$a = 0.324$ $c = 0.5206$
Density	5.606 g/cm^3

neity, which creates different sites for adsorbate to settle down. Usually, the activated carbon is used to remove unwanted materials. The physical compounds which are interacting the number of carbonaceous impurities present, specific chemicals present, and the physical and chemical properties of the activated carbon decide the efficiency and capacity of adsorption of the organic molecules [26]. Dunicz explained the surface area calculation of the activated carbon using Langmuir adsorption isotherm. The interaction between the activated carbon and chemicals is usually π - π interactions due to van der Waal forces or hydrogen bonding [13]. The environmental factors like pH, temperature, the concentration of adsorbate, physical size, ionic concentrations, pore blockage, and diffusion of small compound particles decide the efficiency of adsorption rate of the activated carbon. The activated carbon can be used as a substrate over the metal oxide surface to adsorb the chemical gases present [27].

3. Experimental Procedure

The experiment involves the activated carbon in the ZnO nanowires, which can be used as a sensor for detecting SO_2 . To adsorb more SO_2 , more compound sites are required. The interstiation of SO_2 with the surface requires

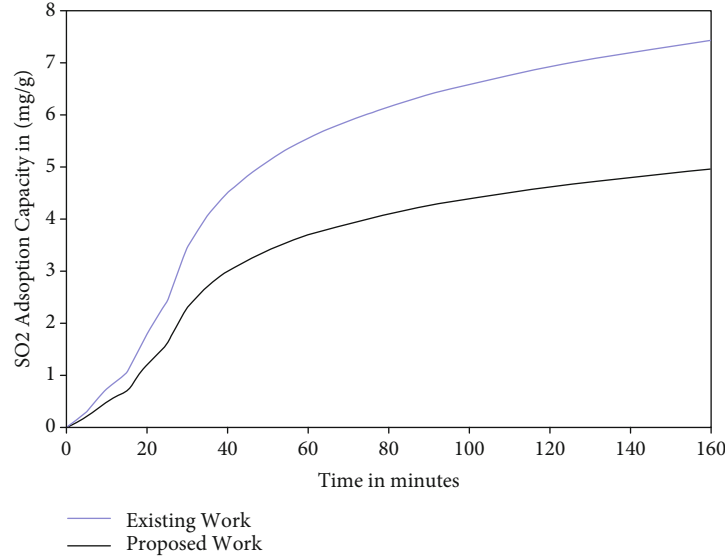
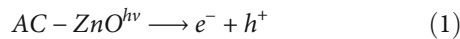


FIGURE 2: Plot of time vs. SO₂ adsorption capacity.

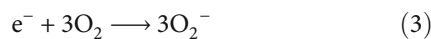
more molecules for bonding. With the help of bonding, more charge transfer has occur, which helps detect the SO₂ presence in the environment. In the case of selecting only the SO₂, there are more charge transfers, which helps avoid other gaseous substances present in the environment and select the SO₂ alone [28]. Both ZnO nanowires and activated carbon have a large surface area from the literature survey. So here, the paper proposes the new method of introducing the activated carbon in the ZnO nanowire substrate to detect SO₂. Few researchers described the usage of the activated carbon for the removal of SO₂. By utilizing the property of a large surface area of the activated carbon, the paper proposes the detection of SO₂ in this work [29]. The low-volume pores of the activated carbon form adsorption sites for the SO₂ adsorbate to settle down on the substrates. The heterogeneity of the activated carbon helps the substrate of ZnO nanowire to produce more cation and anions. In this way, the band excitation occurs by moving electrons from the valence band to the conduction band. The following reaction will take place as shown in equation (1). The photocatalytic equation shows the formation of electrons and holes that will react with the oxygen molecules [30].



When the ZnO adsorbs the SO₂, the following reaction will take place:



Therefore, 3 oxygen molecules formed will react with electrons produced during the AC-ZnO reaction which is explained in



The 3 oxygen ions help the electrons to move from valence band to conduction band, thus by increasing the

conductivity which directly improves the selectivity property of the ZnO nanowire sensor.

The Langmuir adsorption isotherm is used to calculate the capacity of the proposed work. The reason for choosing this isotherm is that it manipulates solid-gas interactions. The isotherm is considering that the adsorbates are gaseous, which get adsorbed to the adsorbent sites of the solid state. The following assumptions are made in the Langmuir adsorption isotherms:

- (I) The immobile state of adsorbate should get adsorbed
- (II) The energy is equivalent in all sites of adsorbent
- (III) The site should have the ability to hold at least one molecule
- (IV) The surface of the adsorbent must be homogeneous
- (V) The interaction of molecules on the adsorbent's adjacent sites should be either zero or ideal

The Langmuir adsorption isotherms formula is given in [14, 17]

$$\theta_A = \frac{K_{eq}^A p^A}{1 + K_{eq}^A p^A}, \quad (4)$$

where θ_A is the fractional occupancy of adsorbent sites, K_{eq}^A is the equilibrium constant, and p^A is the adsorbate's partial pressure.

Another equation described by Langmuir is given in

$$\theta_A = \frac{V}{V_m}, \quad (5)$$

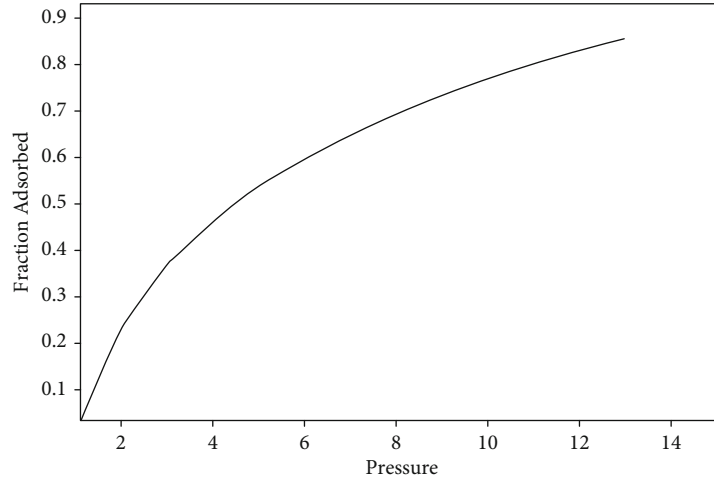


FIGURE 3: Analysis of Langmuir adsorption isotherm.

where V_m is the volume of adsorbate in an immobile gaseous state and V is the volume of homogeneous adsorbent sites where the adsorbates get occupied.

The adsorption of the acid by the activated carbon can be described using Langmuir isotherm [13] as shown in

$$\frac{C}{X} = a + bc, \quad (6)$$

where C is the concentration of SO_2 adsorbed on the adsorbent sites, X is the number of moles adsorbed on the 1 gram of adsorbent, and a and b are the constants.

In equation (6), the value of a will tend to be very small if the adsorbate concentration increases, so the value is omitted.

$$\frac{C}{X} = bc. \quad (7)$$

By taking reciprocal of equation (7), we get

$$X_{\max} = \frac{1}{b}. \quad (8)$$

Equation (8) shows the maximum amount of adsorbate adsorbed on the 1 gram of adsorbent. Therefore, the specific surface area of the adsorbent can be calculated by

$$S = \frac{1}{b} \times N \times 21 \times 10^{-20} \text{ meters}^2, \quad (9)$$

where S is the specific surface area in meters^2 and N is the Avogadro number.

Using equation (9), the adsorbent sites' capacity is calculated for adsorption. The value of b is obtained if the value of S is known already. The value of X_{\max} helps to calculate the amount of adsorbate adsorbed on the various adsorbent sites.

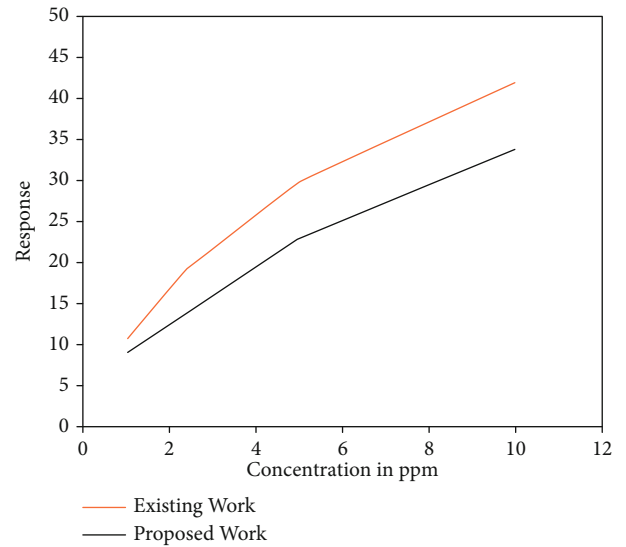


FIGURE 4: Plot of concentration vs. response.

4. Results and Discussions

The experiment for the proposed work is done using MATLAB 2021B software. Table 1 shows the user data for simulation. The ZnO nanowire of size 100 nm is considered. The nominal temperature at 165°C in the simulation is carried out. The assumptions according to the Langmuir adsorption isotherm are made.

Figure 2 shows the plot of the time vs. adsorption capacity of SO_2 on the ZnO nanowire sensor involving activated carbon. The simulation is taken for both normal ZnO nanowire sensor and activated carbon in ZnO nanowire sensor. With the increase in time, it is seen that the adsorption capacity of adsorbate on the adsorbent increases. Due to the addition of the activated carbon in ZnO nanowires, the adsorption rate increases over time. It is seen that the adsorption rate is gradually increasing until an hour. After an hour, the adsorption of SO_2 reaches the saturation point, and it shows a little slope. So it proves that it takes an hour

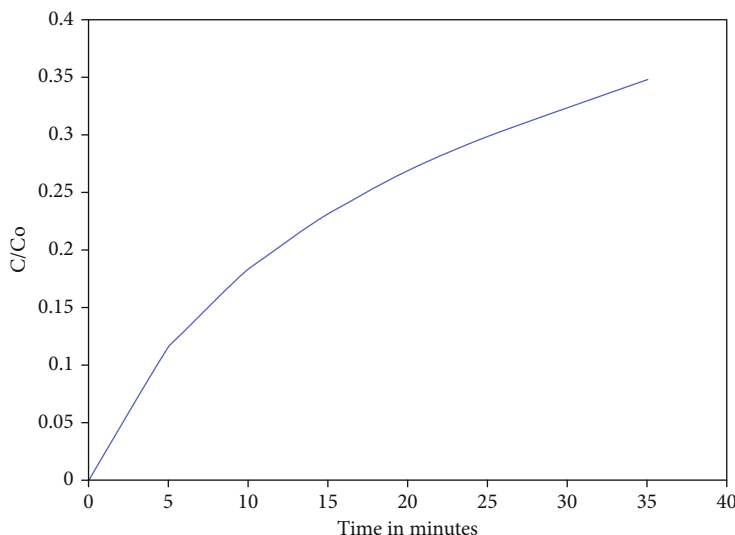


FIGURE 5: Plot of time vs. C/C_0 .

for the adsorbent to start to adsorb the SO_2 adsorbate on the surfaces. From the graphical analysis, the adsorption rate of the proposed work at an hour is 33.34% efficient compared to the existing ZnO nanowire sensor.

Langmuir adsorption isotherm graph is shown in Figure 3. To calculate the solid-gas interaction between the adsorbate SO_2 and the adsorbents of ZnO nanowire using activated carbon, the plot of pressure vs. fractional occupancy on the adsorbent site is done. The graph obtained is similar to the ideal Langmuir adsorption isotherm. Still, some deviations are present. But it shows some of the ideality when compared with it. The values used for simulating this plot are given in Table 1. The ratio of V/V_m reaches almost unity at the end of the process. It means that SO_2 adsorbates occupied all the adsorbent sites. In ratio, the numerator will always be unity but the denominator value will vary from zero to 1. The adsorbate will take some time to get adsorbed on the adsorbent. The adsorbate gradually will get increased, and it reaches the saturation point. From this graph, the proof for SO_2 adsorption capacity which gradually increases with time is also proved.

As the power plants use coal of high ash content, large amount of ash in either form causes severe disposal problems and environmental concern. Their surface disposal causes choking of natural drainage, and the trace elements released from this produce toxicity to animals, plants, and human beings. The bottom ash collected below the combustion unit is discharges from the nearby channel, tailing ponds, or dumpsites. When air-polluting substances leave their site of origin to enter the atmosphere as emissions, they are exposed to many influences before reaching the site, where they take effect as emissions. Sulphur dioxide may drift along with the wind for hundreds of kilometers away from the emission sources. However, large amounts of sulphur dioxide usually get deposited in the areas closer to the sources.

Figure 4 shows the plot of concentration vs. response measured. This calculation is necessary to study the characteristics of the ZnO nanowire sensor response to the SO_2 adsorption. Depending on the concentration of the SO_2 range, the response of the proposed sensor also increases. When the concentration of SO_2 in the environment increases, the response of the sensor detection rate also increases. From the plot, it is clear that the proposed work shows a high response rate compared to existing work. At 5 ppm concentration, the proposed work shows 23% efficiency compared to the normal ZnO nanowire sensor. The response of the sensor mainly depends on the adsorbate present. But at the low amount of adsorbate concentration, even the sensor's response is little good.

The adsorbate and adsorbent concentrations complete each other so that the SO_2 absorbency increases gradually with time. Figure 5 shows the plot of time vs. C/C_0 . The analysis is carried out to find the concentration gradient with an increase in time. There are gradual increases, which shows that the adsorbate gradually settles in the adsorbent sites regarding time. Once the C/C_0 reaches unity, the plot shows the saturation level which is also an interpretation of Langmuir adsorption isotherm.

The particulate matters are biologically active and may have direct toxic effects on themselves, indirect toxic effects through interactions with other pollutants, and chronic effects through cell transformation or regular alternation in cell function. The chemically inert soot emitted by insufficient coal and fuel oil combustion is less dangerous for plants, the particulates from industrial process ($>10\text{pm}$) promptly to other parts of the body.

5. Conclusions

The various simulation paradigms are used to study the proposed work of usage of the activated carbon in the ZnO

nanowire sensor. The adsorption capacity of the sensor increased by 33.34%. The sensor's response to the SO₂ adsorbate increases to about 23%. The novel method follows the isotherm of Langmuir adsorption isotherm. The more adsorbent sites adsorb more SO₂ in the environment, increasing the charge transfer on the surface of the ZnO nanowire sensor. The selectivity of SO₂ compounds from the environment mainly depends on their concentration. The chemical reactions due to adsorption decrease the energy gap and increase the conduction of the ZnO nanowire sensor. When there is more conduction, there increases the electron transfer, which indirectly increases the sensor's signal strength. The increase in the signal strength helps to detect SO₂. The future work of this paper can be improvised by increasing the efficiency of the sensor to detect SO₂ even at lower concentrations in the environment. The carbonaceous impurities of the activated carbon will degrade the sensor's performance. The impurities will react with the oxygen groups in the ZnO nanowire surface. The array of the proposed work will also be used to enhance the selectivity of the SO₂, and the sensitivity to the low concentration rate of SO₂ improved.

Data Availability

The data used to support the findings of this study are included within the article. Should further data or information be required, these are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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