Conductometric gas sensing behavior of WS$_2$ aerogel

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The gas sensing characteristics of porous tungsten disulfide (WS$_2$) aerogel are investigated. The sensor is fabricated by integrating WS$_2$ aerogel onto a low power polycrystalline silicon microheater platform to provide control over the operating temperature. The WS$_2$ aerogel is characterized by scanning electron microscopy, transmission electron microscopy, Raman spectroscopy, X-ray diffraction, X-ray photoelectron spectroscopy, and thermal gravimetric analysis. The sensing performances of the WS$_2$ aerogel-based sensor to NO$_2$, O$_2$, NH$_3$, H$_2$, and humidity are investigated, indicating a p-type behavior. The optimum sensing temperature is found to be about 250°C, when considering sensitivity, power consumption and response time. The role of O$_2$ in the sensor performance is probed and is found to be helpful for enhancing the sensitivity and recovery of the sensor in H$_2$, humidity and NH$_3$.

Introduction

There remains a strong need for miniaturized, low-power gas sensors that can be deployed in wireless applications for improved environmental protection and safety. The detection of gases such as NO$_2$, which is toxic at ppb-level concentrations [1], H$_2$ which is explosive and flammable at 4–75% concentrations in air [2], and NH$_3$ which can result in lung damage and death at high concentrations [3], requires advances in sensing materials to provide the necessary performance characteristics.

Two-dimensional (2D) transition metal dichalcogenides (TMDCs) with lamellar structures similar to that of graphite have received significant attention in recent years. As with graphene, TMDCs can be isolated into two-dimensional sheets, but unlike graphene, they are often semiconductor with sizable band gaps. For example, bulk tungsten disulfide (WS$_2$) is an indirect-bandgap (1.4 eV) semiconductor, but becomes a direct-bandgap (~2.1 eV) material when exfoliated into the monolayer state [4]. More recently, 2D TMDCs are being explored as promising alternatives to graphene for many device applications including gas sensors, due to their high surface-to-volume ratio and adjustable bandgap [5–8].

Single- or few-layer MoS$_2$ has been found to have high sensitivity for NO$_2$, H$_2$, NH$_3$, and triethylamine gas [9–12]. However, limited reports on the gas sensing characteristics of WS$_2$ [4,13–17] are available. Furthermore, the recovery of WS$_2$-based gas sensors is typically incomplete at room temperature because of strong gas adsorption, which limits their practical applications [13,18].

In order to keep the low-power consumption of sensor while heating the sensing material to enhance its response and recovery, a microfabricated heater is needed. However, integration of single-layer materials onto a microfabricated sensor device is difficult, and the surface area is limited by the device footprint. Assembling 2D sheets into 3D aerogel structure provides a new way for enhanced sensing performances by maintaining a high surface area in an accessible porous network [19–22].

Given the potential of aerogels for gas sensing and the related lack of knowledge of the gas sensing properties of WS$_2$, an exploration of the gas sensing capability of WS$_2$ aerogel is in order. Here, we report the first fabrication and characterization of a low-power gas sensor based on WS$_2$ aerogel. The WS$_2$ aerogel is synthesized using thermal decomposition of ammonium thio-metal salts [23]. The aerogel is integrated onto a low-power silicon microheater platform, in order to control the operating temperature and enhance the...
sensing response and recovery of the aerogel. The sensing behaviors of the WS$_2$ aerogel to NO$_2$, O$_2$, H$_2$, NH$_3$, and humidity in dry air and N$_2$ are reported, including the sensor’s dependence on temperature. The role of background oxygen in sensing performances is also reported and a possible gas sensing mechanism is proposed.

Experimental

Synthesis and characterization

WS$_2$ aerogel synthesis follows the procedure reported in Ref. [23]. Briefly, 36 mg of ammonium thio-tungstate (ATT) is dissolved in 1 mL of deionized water in a vial. The vial is then subjected to a freeze-drying cycle to produce an ATT aerogel. The ATT aerogel is annealed in 2% H$_2$/98% Ar mixture at 750°C for 1 h to yield the WS$_2$ aerogel.

WS$_2$ aerogel is characterized using scanning electron microscopy (SEM, FEI Sirion XL30) and transmission electron microscopy (TEM, JEOL 2010). Raman spectroscopy (HORIBA Jobin Yvon system equipped with an Olympus microscope, and a 632.8 nm laser as the excitation source), X-ray diffraction (XRD, AXS D8 Discover General Area Detector Diffraction System form Bruker with radiation from a Cu target, $\lambda = 0.15406$ nm), and X-ray photoelectron spectroscopy (XPS, Omicron Dar400 system with an achromatic Al K$_\alpha$ X-ray source) are used for elemental and structural analyses.

Sensor fabrication and sensing testing

The details of the microheater fabrication process can be found in a previous report [24]. A silicon substrate is coated with silicon-rich low-stress silicon nitride (100 nm) followed by the deposition of phosphorus-doped polysilicon (100 nm), which is patterned using photolithography to form the microheater. Another silicon nitride layer (100 nm) is deposited and patterned for the microheater contacts, which are electron-beam evaporated Ti/Pt (10/90 nm). The back side of the wafer is patterned and etched using KOH to release the silicon nitride membrane. The Ti/Pt electrodes overlaid on the silicon nitride membrane are used as electrodes for conductometric sensing.

Conductometric gas sensors are prepared by drop casting a suspension of WS$_2$ aerogel material onto a low-power microfabricated heater platform. The WS$_2$ aerogel is dispersed in isopropyl alcohol (0.5 mg/mL) by sonication and drop cast (five drops of 0.2 $\mu$L each) onto the microheater chip while the microheater is heated to 200°C and maintained there for 3 h to promote solvent evaporation and material deposition at the center of the microheater.

For sensor testing, the sensor is placed in a gas flow chamber ($\sim$1 cm$^3$ volume). Gas exposure and signal collection are controlled using Labview and an open-source Java-based instrument and control and measurement software suite, Zephyr. Gas tanks of O$_2$ (Praxair, 99.995%), H$_2$ (Praxair, 5% in N$_2$), NO$_2$ (Praxair, 20 ppm in N$_2$), and NH$_3$ (Praxair, 100 ppm in N$_2$) are controlled with mass flow controllers (Bronkhorst) and mixed with N$_2$ or dry air to give the desired gas concentration with a total flow rate of 300 sccm. Relative humidity (RH) is adjusted by evaporating deionized water and mixing it with gases. For sensor signal collection, a bias voltage is applied to the microheater and the microheater resistance is measured with a Keithley 2602A source-meter.

Results and discussion

Characterization methods

Each single layer of WS$_2$ comprises of a tungsten layer sandwiched between two sulfur layers in a trigonal prismatic coordina-

ation as shown schematically in Fig. 1a. Fig. S1 (in Supporting Information) shows an optical image of as-synthesized WS$_2$ aerogel. Scanning electron microscopy images in Fig. 1b–c show the connected nanosheet structure of the WS$_2$ aerogel. The aerogel density is 35 mg/cm$^3$, which is 0.5% of the bulk density of WS$_2$, with a surface area of 9 m$^2$/g calculated from nitrogen adsorption analysis [23] (Supporting Information, Fig. S2). After deposition on the microheater device, the sensing material maintains the porosity and the connected nanosheets of the aerogel (as shown in Fig. S1, Supporting Information), as judged by comparing the material on SEM stub and microheater device after deposition. Fig. 1d shows a representative low-magnification TEM image of WS$_2$ stacked nanosheets. The measured spacing between the fringes in the lattice is approximately 6.1 Å (Fig. 1e), which matches well the (0 0 2) d-spacing for the layered WS$_2$ structure. The high-resolution TEM image in Fig. 1f shows Moiré fringes indicating that the WS$_2$ sheets contain many layers, with thicknesses between 2 and 8 nm (Fig. S3 in Supporting Information, and Fig. 1f).

To confirm the structure and composition of the aerogel, Raman spectroscopy is performed. The WS$_2$ Raman spectrum is characterized by the first order modes at the Brillouin zone center, known as $E_{2g}$ and $A_{1g}$. Previous studies on the Raman spectroscopy of WS$_2$ report that the $A_{1g}$ mode blue shifts with an increasing number of layers, while the $E_{2g}$ mode exhibits a redshift [25–27], with the $A_{1g}$ and $E_{2g}$ peaks of monolayer WS$_2$ reported at 417 and 355 cm$^{-1}$, respectively [25]. Fig. 2a shows that the major peaks found for the WS$_2$ aerogel are at 420 cm$^{-1}$ (A$_{1g}$) and 350 cm$^{-1}$ (E$_{2g}$). Hence, the WS$_2$ aerogel used in the present study is composed of multiple stacked layers, consistent with the TEM analysis of the material.

X-ray photoelectron spectroscopy is performed to determine the oxidation states of the W in the aerogel. In Fig. 2b, the peaks located at 33.1, 35.2, and 38.6 eV correspond to W 4f$_{7/2}$, 4f$_{5/2}$, and W 5p$_{3/2}$ binding energies (BE), respectively. The energy positions of these peaks indicate a W valence state of +4, which is in accordance with the previous reports, indicating the formation of pure WS$_2$ phase [28]. At the same time, nearly all the S 2p signal is from S$^{2−}$ (2p$_{3/2}$, BE = 162.6 eV; 2p$_{1/2}$, BE = 163.7 eV), also typical of WS$_2$. The sulfur to tungsten ratio is calculated to be 2.05. The analysis also indicates the absence of any WO$_3$ in the aerogel.

The X-ray diffraction pattern (Fig. 2c) shows characteristic peaks for WS$_2$ at $2\theta = 14.2°$, 28.5°, 33.3°, 39.5°, 43.3°, and 58.5°, which respectively correspond to the (0 0 2), (0 0 4), (1 0 0), (1 0 3), (1 1 0), and (2 0 0) planes of hexagonal 2H-WS$_2$ (JCPDS-ICDD 71-4832). The (0 0 2) peak suggests that the turbostratic stacking is dominant in the aerogel [29]. The broadness of the peaks is indicative of the randomly connected nanostructures that make up the aerogel. This result is consistent with the SEM and TEM images (Fig. 1b–f).

Thermogravimetric analysis (TGA) performed under 20% O$_2$ and Ar mixed gas is presented in Fig. 2. The aerogel shows some weight loss at temperatures below 100°C, which is attributed to the removal of adsorbed water. It begins to degrade to form WO$_3$ at around 400°C, with a final weight loss of 6.5%, which is consistent with the full conversion of WS$_2$ to WO$_3$. Therefore, sensing tests must be performed at temperatures below 400°C to ensure that the WS$_2$ is not oxidizing during testing.

Gas sensing performances

The microheater used in this work can reach 300°C with only 5 mW peak power. The schematic diagram of the sensor is shown in Fig. 3a, indicating the suspended microheater and the WS$_2$ aerogel deposited above the heater. An optical image of the microheater chip (3.5 × 3.5 mm$^2$) containing four microheater sensors

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is shown in Fig. 3b. Each microheater sensor has four electrical contacts: two for the microheater leads and two for electrical probing of the sensing layer. Fig. 3c shows a zoomed-in view of a single microheater sensor showing the polysilicon heater, the silicon nitride membrane and the Ti/Pt sensor leads. Fig. 3d shows the power input to the microheater versus temperature of the microheater, which is calibrated by measuring the radiation spectrum with the heater powered to the glow point and fitting it to Planck's distribution, as described in Ref. [30].

The current vs. voltage characteristics across the WS$_2$ aerogel (Fig. 4a) display a nearly linear behavior in the temperatures range from 20 to 300 °C, suggesting the formation of an Ohmic contact with the Pt/Ti sensing electrodes. Fig. 4b shows a decreasing resistance with temperature increasing, consistent with the typical behavior for semiconductors. Namely, as the temperature increases, carriers are thermally activated and the resistance decreases following an exponential relationship, $R(T) = R_0e^{(E/kT)}$, where $E$ is the activation energy and $k$ is the Boltzmann constant. The thermal-activation model yields a good fit to the dataset as shown in Fig. 4b, with an activation energy of 0.30 eV. This behavior is in agreement with previous report on WS$_2$ nanowires [31].

For gas sensing measurements, the sensor voltage is maintained at 1 V. The gas sensitivity is calculated as $\Delta R/R_0 = (R_0 - R_{gas})/R_0$, where $R_0$ and $R_{gas}$ represent the resistance of the device to the background gas and the analyte gas, respectively. The response time ($t_{response}$) is defined as the time taken to reach 90% of the full response after the introduction of the target gas. The recovery time ($t_{recovery}$) is defined as the time taken to return to 90% of the baseline resistance after the flow of target gas is stopped.

Fig. 5a shows the sensor response to 2 ppm NO$_2$ in pure N$_2$ at 20 °C for 15 min duration pulses. The Fig. 5a indicates that the WS$_2$ aerogel can detect NO$_2$ gas at room temperature, although the response does not stabilize and the rate of signal recovery is very low. Fig. 5b shows the temperature dependence of the response to 2 ppm NO$_2$ in pure N$_2$, with the temperature ranging from 20 to 300 °C. There is a clear trend from slower response time and incomplete recovery at room temperature to faster response and recovery at higher temperatures, with a similar recovery behavior at 250 and 300 °C. Additionally, the response rate and sensitivity appear to increase with increasing temperature, although the sensitivity at low temperatures is difficult to quantify when the sensor does not reach a stable value. The sensing behavior versus temperature is similar to the response and recovery results for other TMDCs-based gas sensors [32,33]. Considering the improved response and recovery to NO$_2$, the thermal stability of the WS$_2$ aerogel, and the power consumption of the sensor, the heater temperature of 250 °C is considered the optimum operating temperature. Further sensing tests are taken at 250 °C to facilitate comparison of the response to other gases.

The NO$_2$ exposure tests reported in Fig. 5 were performed in nitrogen, but in practical applications, the sensor is exposed to air. In order to determine whether this has an impact on the sensor characteristics, the WS$_2$ aerogel sensor is exposed to oxygen at varied concentrations. The resistance of the WS$_2$ aerogel sensor versus time to different concentrations of O$_2$ in N$_2$ ranging from 500 to 5000 ppm with the heater at 250 °C was tested. Fig. 6 shows the response increasing monotonically with O$_2$ concentration in this range. The resistance of a WS$_2$ aerogel sample decreases upon exposure to O$_2$, and increases when the O$_2$ flow is turned off. For
sensors operating in air, this indicates that the sensor baseline will be lower than expected. It also suggests the importance of carefully controlling the oxygen concentration during exposure tests balanced in air such that any sensor response is attributed to the change in the target gas concentration, and not oxygen concentration.

Theoretical calculations and experimental results on the gas sensing characteristics of graphene and MoS2-based devices have...
demonstrated high sensitivity to various gases due to the charge transfer between target gases and sensing materials, where oxidizing gases act as electron acceptors, and reducing gases are electron donors [12,34,35]. In our case, when the WS2 aerogel sensor device is exposed to oxidizing gases (NO2 and O2), the observed decrease in the sensor resistance suggests p-type behavior, which is in agreement with some previous reports [36–38]. McDonnell et al. [39] demonstrated systematically by experiments that sulfur-deficient and sulfur-rich MoS2 would be expected to result in n-type and p-type behavior respectively. Probably, a somewhat rich S from the precursor (ammonium thiotungstate) results in the p-type behavior of WS2 aerogel. This is consistent with the XPS spectra (Fig. 2f), indicating S-rich WS2.

The sensor response to varied concentrations of NO2 from 0.2 to 3 ppm in dry air is shown in Fig. 7a with the heater at 250 °C. The resistance of the sensor decreases upon exposure to NO2, consistent with p-type behavior. Fig. 7b shows the response of the sensor increasing with increasing NO2 concentration, saturating above 2 ppm. Using the baseline noise level and the conventional signal-to-noise ratio of 3 implies a limit of detection of 8 ppb for NO2, which is well below the 100 ppb threshold of interest [1]. The response time decreases from 6 to 2 min with increasing NO2 concentration while the recovery time ranges from 10 to 30 min.

![Fig. 4](image1.png)

**Fig. 4.** (a) I/V characteristics of the WS2 aerogel sensor at different temperatures from 20 to 300 °C. (b) Typical sensor resistance vs. temperatures with a sensor voltage of 1 V.

![Fig. 5](image2.png)

**Fig. 5.** (a) Response of the WS2 aerogel sensor to three cycles of 2 ppm NO2 in pure N2 at 20 °C. (b) Response of the sensor to 2 ppm NO2 in pure N2 at different temperatures (20, 100, 200, 250, 300 °C) showing improved response and recovery times with increasing temperature.

![Fig. 6](image3.png)

**Fig. 6.** Sensitivity of the WS2 aerogel sample to 500–5000 ppm O2 in pure N2 at 250 °C. The error bars represent the standard deviation of response of the 5 exposures to a given O2 concentration.
For the typical reducing gas H₂, the resistance of the sensor increases upon exposure to H₂ with the heater at 250 °C demonstrating the p-type of WS₂ aerogel, as shown in Fig. 7c. Fig. 7d shows the response increasing monotonically with H₂ concentration in the range of 50–5000 ppm. The signal-to-noise ratio of 3 yields a limit of detection of 60 ppm for H₂, which is well below the lower explosive limit of H₂ (4000 ppm)[2]. The response and recovery times range from 1 to 2 min for all hydrogen concentrations.

For NH₃, another reducing gas, the resistance of the sensor also increases upon exposure to NH₃ with the heater at 250 °C (Fig. S5, Supporting Information). The sensitivity increases monotonically with NH₃ concentration in the range of 50–800 ppm. The signal-to-noise ratio of 3 implies a limit of detection of 13 ppm for NH₃[3]. The response and recovery times range from 3 to 5 min for all NH₃ concentrations. Compared to most TMDC-based sensors, which do not reach steady state upon exposure to NO₂ or NH₃, nor exhibit complete[13,34,40], the WS₂ aerogel sensor reaches steady state with over 90% recovery in the examined concentration range at microheater temperature of 250 °C. Furthermore, the WS₂ aerogel is highly stable. After extended gas sensing measurements at 250 °C, the WS₂ remains unchanged, according to the Raman spectra before and after one week of sensing tests at 250 °C (Fig. S7, Supporting Information).

The sensitivity values to H₂ and NH₃ are much lower than NO₂ given that the responses are comparable but for very different concentration ranges (0.2–3 ppm for NO₂ versus 500–5000 ppm for H₂ and 50–800 ppm NH₃). The larger adsorption energy of NO₂, compared to that of H₂ and NH₃, may be responsible for the higher sensitivity and longer response/recovery time of the sensor in NO₂[41], as has been proposed earlier in the case of graphene-based sensors[42,43].

Because the WS₂ aerogel sensor is affected by the presence of oxygen as shown in Fig. 6, the role of oxygen in the sensing performances of this sensor is further elucidated. In Fig. 8a, the sensor is exposed to 2000 ppm H₂ with varied O₂ content between 0 and 21% balanced in dry N₂. Fig. 8b-c show the sensitivity and response/recovery times as a function of O₂ content in N₂. The sensor resistance increases upon exposure to H₂ in all measurement atmospheres (Fig. 8a), which is in agreement with Fig. 7c, while the baseline resistance of the sample decreases with increasing O₂ content in N₂, as expected from Fig. 6. However, the existence of oxygen, in particular in 5–21% range, sharply enhances the sensitivity of the WS₂ aerogel. Increasing the O₂ content continuously improves the recovery time, while it has no obvious effect on the response time.

Similarly, O₂ enhances the sensing performances of WS₂ aerogel to humidity and NH₃, as shown in Fig. 8d-e. Clearly, recovery of the sensor is incomplete in pure N₂, while the sensor recovers well in dry air. Sensitivity values are increased and response time values are decreased to both humidity and NH₃ in dry air (see Supporting Information Table 1).

Although the majority of recent reports conclude that the primary response mechanism for TMDC gas sensors is charge transfer between the adsorbed gas and the sensing material, our results indicate that the effects of adsorbed oxygen cannot be ignored. The weak response to reducing gases (H₂, humidity and NH₃) in pure N₂ is consistent with theoretical studies that predict a low

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**Figure 7.** (a) Resistance versus time and (b) the sensitivity of the sensor to NO₂ in dry air with the microheater at 250 °C. (c) Response versus time and (d) the related sensitivity of the sensor to 500–5000 ppm H₂ in dry air with the microheater at 250 °C. The error bars represent the standard deviation of response of the 5 exposures to a given NO₂ (or H₂) concentration.
degree of charge transfer between these three molecules and WS2 [16]. The presence of oxygen in dry air further enhances the sensitivity of WS2 to H2, humidity and NH3. Similar behavior is reported on horizontally aligned carbon nanotubes for H2 sensing [44] and liquid-exfoliated MoS2 for ethanol sensing [45], and is attributed to the adsorbed oxygen promoting the interaction kinetics between the reducing gases and the sensing materials. On the contrary, investigation of NO2 sensing in pure N2 compared to in dry air shows a small effect of O2 (Supporting Information, Fig. S6). This is most likely because the degree of charge transfer for NO2 is so strong that WS2 resistance is unaffected by O2 presence, or NO2 has no interaction with adsorbed O2 molecules.

Although a few conductometric gas sensors based on WS2 have been reported previously [4,13–17], limited information is available on the selectivity and NO2 sensing performance of these sensors. The comparisons for WS2-based gas sensors are listed in Table 1. The WS2 aerogel-based sensor, in present work, shows p-type characteristics, allowing easy differentiation of oxidizing and reducing gases in air as shown in Fig. 8f, as well as excellent NO2 sensing performance with great selectivity in N2. This behavior is different from gas sensors based on metal oxides (such as WO3), working in rich oxygen environment [47]. Compared to previous WS2 nanosheets-based NO2 sensor [46], the aerogel-based sensor here shows short recovery time to low concentrations of NO2.

Conclusions

We have experimentally demonstrated a low-power microsensor utilizing WS2 aerogel sensing material. The response of the sensor to NO2, O2, H2, humidity, and NH3 were investigated in dry air and N2 respectively. Increasing the operating temperature to 250 °C (<5 mW) results in both increased sensitivity and shorter response and recovery time. Due to larger adsorption energy of NO2, WS2 aerogel shows stronger response to NO2 than to other gases with longer response and recovery time. O2 enhances the sensing performances of the sensor to reducing gases (H2 and NH3) and humidity, while it has a very weak effect on NO2 sensing. The WS2 aerogel is promising for NO2 sensing in low or even without oxygen environment.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.flatc.2017.08.003.

References


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