Introduction

1st paragraph

However, due to its inherent non-selectivity, and huge p-doping in atmospheric air, it's applications in gas sensing are often limited to controlled environments such as nitrogen, dry air, or synthetic humid air.

P-doping is a process of introducing impurities or dopants that create excess holes in a semiconductor material, making it a p-type semiconductor.

Van der Waals forces are weak intermolecular forces that are dependent on the distance between atoms or molecules. These forces arise from the interactions between uncharged atoms/molecules.

2nd paragraph

The CNPD method, or disparity measurement, is a technique used to measure the interaction between graphene and adsorbed gas molecules. The procedure involves applying a constant voltage (Vt) to the graphene channel gate electrode to tune the van der Waals (vdW) interaction between the graphene and the adsorbed gas molecules. The changes in the interaction are then monitored by transfer characteristics measurements immediately after the voltage application.

This sequence of Vt application followed by transfer characteristics measurement is repeated several times. The difference between the charge neutrality points (CNP) from successive transfer characteristics curves is obtained as a measure of the effect of the tuning voltage-induced change in the graphene-gas vdW complex charge transfer, referred to as the CNPD values.

The disparity measurement method effectively tracks the electric field modulated charge transfer response of the graphene-molecule vdW complex by harnessing the signature charge transfer of the adsorbed gases, the van der Waals bonding memory of

the gas adsorption induced graphene-molecule vdW complex, and the real-time switching of the vdW bonding between various electrically tuned states in real time.

However, the method is time-consuming, requiring 30-45 minutes measurement time for an environment. Additionally, similar gas environments, such as atmospheric gases, often show overlapping CNPD values, making delineation of complex gas environments subjective.

In other words
The CNPD method is a technique that measures how gas molecules stick to graphene. It involves putting a voltage on the graphene to change how it interacts with the gas, and then measuring how the gas molecules stick to the graphene. This process is repeated several times, and the difference between the measurements is used to calculate the effect of the voltage on the interaction between the graphene and the gas.

This method is good for tracking how the interaction changes in real-time, but it takes a long time to do and can be hard to tell the difference between similar types of gas.

3rd paragraph

Scientists have been using a technique called machine learning to identify different types of gas using just one device. However, the success of this technique depends on the type of information, or "input features," that the machine learning model is given.

Traditional machine learning models for gas detection only look at how gases change the way electricity flows through a material (like graphene). They don't take into account other factors that might affect how gases interact with the material, such as the application of a voltage.

A new technique called the disparity measurement method allows scientists to see how gases interact with a material (graphene) before, during, and after a voltage is

applied. This can provide more detailed information that could help machine learning models better identify different gases, especially in complex gas environments like the air we breathe.

In the context of the passage, "input features" refers to the different types of information that are used to train a machine learning model to identify different gases. For example, in the passage, the input features for traditional graphene-based machine learning models are limited to only monitoring changes in the electrical conductivity of graphene induced by different gases. In contrast, the disparity measurement method allows for additional input features related to how gases interact with graphene in response to applied voltages, which could provide more detailed information and potentially improve the accuracy of machine learning models.

The disparity measurement method is a technique for monitoring the interaction between graphene and adsorbed gas molecules, specifically the van der Waals (vdW) complex formed between graphene and the adsorbed gas.

Understanding: The disparity measurement method itself is not used for gas identification. Rather, it is used to monitor the interaction between graphene and different gas molecules in real-time by measuring the changes in charge transfer induced by the vdW complex formed between the graphene and the adsorbed gas molecules.

The researchers in this study used the data obtained from the disparity measurement method as input features to train machine learning models to identify different gas molecules. These input features were used in combination with other features such as mobility response, charged impurity concentration, and others. The models' performance was dependent on the selectivity of the input features, meaning that some input features were better than others for identifying certain gas molecules.

In summary, the disparity measurement method is a technique for monitoring the interaction between graphene and different gas molecules in real-time. It provides data that can be used as input features for machine learning models to identify different gas molecules. However, the actual identification is done by the machine learning models, not the disparity measurement method itself.

Yes, that's correct. The charge transfer between graphene and the adsorbed gas molecules, which is monitored using the disparity measurement method, can be a key factor used to identify different gas molecules. Different gas molecules can have different effects on the charge transfer, and these differences can be used as input features for machine learning models to distinguish between them.

4th paragraph

In this work, a machine learning model was developed to identify atmospheric gases using a sensor called a-CF-GFET. The sensor was designed to make graphene interact better with gases in the air. The model used eight different features to monitor how the graphene-gas interaction changed when a voltage was applied. The models were tested in different experimental conditions and could accurately distinguish between different gases in the air with 100% accuracy. The contribution of each feature to the model's predictions was also explained using a method called SHAP.

CF-GFET stands for Carbon-based Field-Effect Transistor with a Graphene Channel. It is a type of sensor that can detect changes in the electrical conductivity of graphene caused by the presence of gas molecules. By using a-CF-GFET, which is a modified version of this sensor, the researchers were able to achieve better sensitivity and selectivity for gas detection in atmospheric environments.

Materials and Methods

A) Experimental Section

The device used in the study was created by putting a layer of graphene, which was made through a process called chemical vapor deposition, on a copper substrate. This layer of graphene was then transferred onto a substrate made of silicon and silicon dioxide using a solution of ammonium persulfate. The device was then heated in a mixture of argon and hydrogen gases at 300 degrees Celsius for 3 hours to improve its properties.

Afterwards, small metal electrodes made of chromium and gold were added using a process called electron beam lithography, which involves using a beam of electrons to create patterns on a surface. The activated carbon-functionalized graphene channel, which is where the gas sensing takes place, was also created using electron beam lithography. The channel was first covered with a special kind of resist material and then exposed to an oxygen plasma, which etched away parts of the graphene layer to create the desired pattern. Finally, the resist material was heated to turn it into activated carbon, which is more effective at sensing gases.

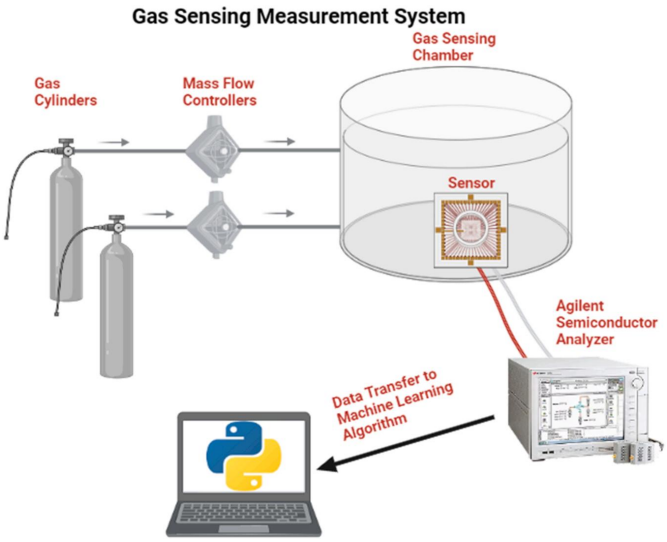
All of these steps together make up the process of device fabrication, which involves creating a device from scratch using various materials and techniques.

The designed pattern is created on the substrate to define the channel through which the gas flows and interacts with the graphene. This pattern also helps to create the electrodes that will measure the voltage changes caused by the interaction of gas molecules with the graphene surface. By designing a specific pattern, the researchers can control the geometry and size of the channel, which can affect the sensitivity and selectivity of the gas sensor. Therefore, the pattern is a critical element in the fabrication process of the gas sensor device.

B) Gas Sensing Experiments

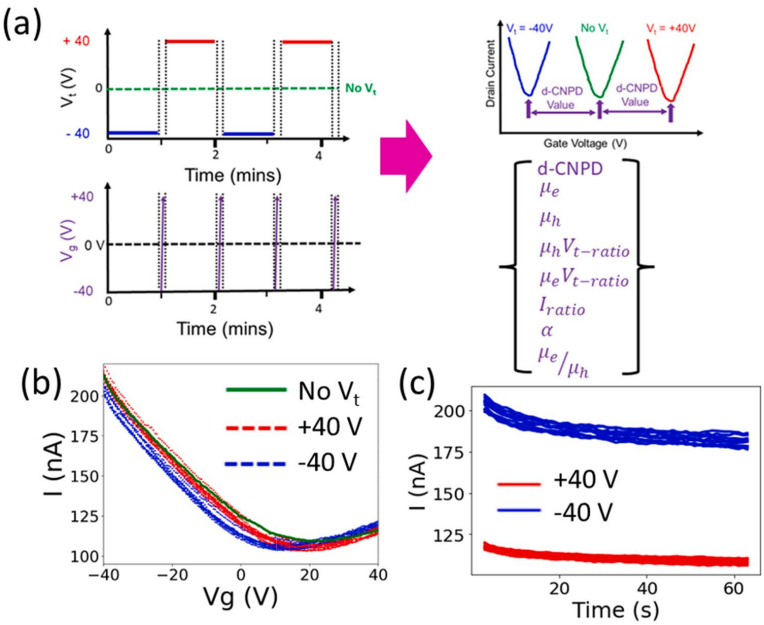
This passage is discussing how the gas sensing experiments were conducted. The researchers used a special device that measures the electrical current flowing through a graphene channel when gases are present. They measured the current under different conditions, including in vacuum, in atmospheric air, in nitrogen, and in dry air. They also introduced different gases at different concentrations into the chamber using gas

cylinders and mass flow controllers. The gases included ammonia and acetone, which were introduced at concentrations ranging from very low (1 part per billion) to relatively high (10,000 parts per million). The researchers also measured the humidity and temperature of the chamber. The goal was to test how well the device could detect different gases under different conditions.



Gas sensing schematic showing the measurement set-up for the a-CF-GFET sensor.

C) Disparity Measurement



Disparity measurement schematic showing the extracted ML features (lower right). It is important to note that the relative positions of the No Vt and + 40 Vt varies with the gas environment and also during a tuning experiment depending on the doping charac teristics of the gas environments at + 40 Vt.

This is evident in the summarized box plot in Fig. S5(a) supporting information. b) Transfer characteristics measurement for 1 ppb ammonia in atmospheric air before Vt (green), after − 40 V (blue), and after + 40 V (red). c) Corresponding drain-source current vs time response of the graphene-molecule vdW com plex for − 40 V (blue), and + 40 V (red line).

The disparity measurement is a way to check the gas sensing performance of the device. First, a voltage is applied to the device to tune the interaction between the graphene and gas molecules for a fixed time. Then, the device is measured to see how it responds to gas molecules. This is done twice with opposite voltages to see if the device responds consistently. This process is repeated several times with short time

lags between each step. The gate voltage range is kept within a certain limit during the entire process. This helps to evaluate the gas sensing ability of the device.

When a gas molecule interacts with the graphene surface, it can donate or accept electrons, creating either a positive or negative charge on the graphene. This, in turn, changes the electrical properties of the graphene, such as its conductivity. This change in conductivity is the basis for detecting the presence of a gas molecule.

However, the interaction between gas molecules and the graphene surface can be affected by various factors, such as the temperature, humidity, and the presence of other gas molecules. Tuning the gate voltage helps to optimize the interaction between the gas molecules and the graphene surface, thereby improving the sensitivity and selectivity of the gas sensor.

Tuning is used to adjust the interaction between the graphene and the gas molecules in order to optimize the gas sensing properties of the device. The gas molecules can affect the conductivity of the graphene by either increasing or decreasing it, depending on the nature of the gas and the graphene surface. By tuning the gate voltage, the interaction between the graphene and the gas molecules can be adjusted, which in turn can improve the sensitivity and selectivity of the gas sensor.

Adjusting means changing or altering something to achieve a desired outcome or to make it better suited for a particular purpose. It could involve making small changes or fine-tuning to improve the performance, accuracy, or effectiveness of a system, process, or device. In the context of the gas sensing experiments described earlier, adjusting the interaction between the graphene and gas molecules means modifying the electrical properties of the graphene to make it more sensitive to the presence of gas molecules, while minimizing the effect of the gas on the graphene's conductivity.

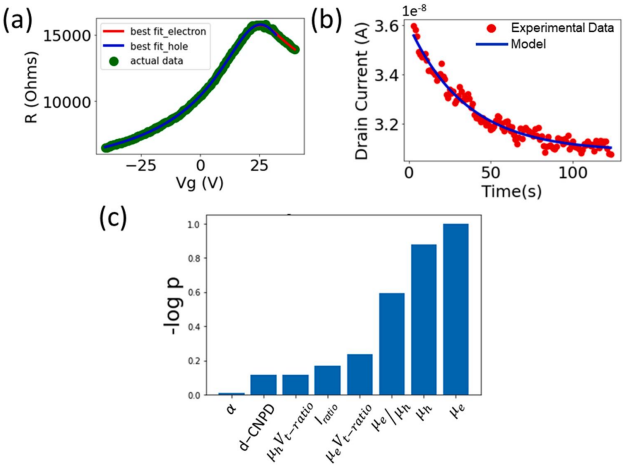
That's correct. Tuning is not done to protect the electrical conductivity of graphene, but rather to enhance its sensitivity to the presence of specific gas molecules. The goal of tuning is to adjust the interaction between the graphene and the gas molecules so that any changes in the electrical properties of the graphene can be correlated with the presence and concentration of the target gas.

In this experiment, the researchers measured the interaction between graphene and different gases (acetone and ammonia) by applying a voltage to the graphene and then measuring its electrical properties. They used a specific sequence of voltage application and measurement, which was repeated several times. The time between each measurement was kept very short, and the range of the voltage applied was limited.

After the gas sensing experiments, the researchers measured the recovery of the graphene device by heating it to high temperature or vacuuming it. They found that the device recovered more quickly in dry air or nitrogen compared to atmospheric air.

Overall, they performed 574 sets of measurements, including measurements of acetone and ammonia in atmospheric air, nitrogen, and dry air, as well as blank atmospheric air without any gases. The researchers emphasized the importance of keeping the tuning time and voltage consistent during the measurements to ensure accurate results.

D) Dataset creation for ML model



a) Electron (red), hole (blue) transport model fitting to Eq. 1, and the raw measurement data (green) for atmospheric acetone after Vt application. b) Drain source current response for atmospheric acetone. c) One way ANOVA analysis of the features, summarized as the normalized negative logarithm of the obtained p-values.

The paragraph explains the methodology used in a research work to study the interaction of gas molecules with graphene. The researchers studied the electron and hole transport regimes separately to obtain the charge neutrality point (CNP) of the gas environment using an equation that includes the sensor contact resistance and sensor channel resistance, number of squares of the gated area, electronic charge, and mobility. The researchers also extracted six ML features from the transfer characteristics curves to track the graphene-gas interaction before and after the application of Vt, including the relative changes in electron and hole mobilities, the effect of Vt polarity on the graphene-gas hole scattering, and the Vt modulated gas adsorption induced charge transfer. The researchers used the drain-source current vs time plot to monitor the gas adsorption induced doping and scattering events in the graphene channel. The conductivity of the graphene channel is a product of the carrier mobility, carrier concentration, and electronic charge.

Results and discussions

1st paragraph

When gas molecules stick to graphene, two things happen:

Some of the electrons from the gas transfer to the graphene, which changes how many electrons are moving through it.

The gas molecules also create an obstacle that slows down the electrons moving through the graphene.

These two effects change how well the graphene can conduct electricity. This can be measured by looking at how much current flows through the graphene. The equation ($\sigma = ne\mu = Ids.L\ Vds.W$) helps explain how these effects influence the amount of current that flows through the graphene.

2nd paragraph

When gas molecules come in contact with graphene, they can either increase the number of electron carriers or hole carriers in the graphene, depending on the type of gas. If the gas-induced scattering events are not significant, then the increase in the number of carriers will result in an increase in conductivity and drain-source current. However, if electron-hole recombination occurs due to the presence of gas, then the total carrier concentration will decrease, and the conductivity and drain-source current will decrease. The application of a voltage can also affect the number of carriers in the graphene, resulting in changes in conductivity and drain-source current. If carrier transport is doping dominated, the adsorption of p-doping gases will increase the drain-source current, while scattering dominated transport will decrease the drain-source current. On the other hand, n-doping gases will decrease the drain-source current, regardless of whether the transport is doping or scattering dominated.

In the context of the passage you provided, scattering events refer to the phenomenon of gas molecules adsorbing onto graphene and creating a scattering potential around the adsorption region. This results in a change in the carrier mobility of the graphene channel, which can affect the conductivity and drain-source current of the device. Essentially, scattering events are instances where the motion of a particle (in this case, the electrons in the graphene channel) is altered by interactions with other particles (in this case, gas molecules). These interactions can affect the behavior and properties of the system as a whole.

In the context of electronics and semiconductors, the term "carrier" typically refers to either an electron or a hole that is free to move within a material and carry an electric charge. In a semiconductor device, such as a graphene-based sensor, the concentration and mobility of carriers play a crucial role in determining its electrical conductivity and response to external stimuli.

Drain-source current (Ids) is the electrical current that flows through a transistor between its two terminals: the drain and the source. In a field-effect transistor (FET), which is the type of transistor typically used in graphene-based sensors, the drain-source current is controlled by the voltage applied to the gate electrode, which modulates the conductivity of the channel region between the source and drain. The

magnitude of the drain-source current is determined by the conductivity of the channel and the applied voltage.

In electronics, the "drain" refers to one of the three terminals of a field-effect transistor (FET), a type of transistor commonly used in electronic circuits. The drain is the terminal that collects the output current of the transistor. In other words, it's the electrode through which the current flows out of the transistor and into a load, such as a resistor or another transistor. The other two terminals of an FET are the "source" and the "gate."

3rd paragraph

The researchers observed that in both p-doping and n-doping environments, the drain-source current of the graphene-molecule van der Waals complex decreased over time when a negative voltage was applied. This suggests that carrier transport is mainly affected by scattering, rather than doping. When carrier transport is mainly doping-dominated, n-dopants (such as ammonia in dry air) are expected to increase the drain-source current over time, while p-dopants (such as atmospheric air) are expected to decrease it due to electron-hole recombination. However, the researchers found that the drain-source current continuously decreased over time regardless of the gas environment. This indicates that the gas adsorption events were scattering-dominated. These results suggest that adding features to the machine learning model that are sensitive to scattering events could improve its accuracy in predicting gas adsorption events.

Q-doping and n-doping are the processes of intentionally introducing impurities into a material, such as a semiconductor or graphene, to increase the concentration of either positive charges (holes) or negative charges (electrons), respectively. This can be achieved by introducing impurities that have an excess of electrons (n-doping) or by removing electrons (p-doping). P-doping increases the concentration of holes, while n-doping increases the concentration of electrons. The type of doping used depends on the desired electrical properties of the material, such as conductivity or resistance.

4th paragraph

The figure 4a-c and 5a-c shows the different clusters of data corresponding to various environments. Three types of machine learning models (XGBoost, K-nearest neighbors, and Naive Bayes) were used to classify the various gas environments using eight machine learning features. The data was processed by labeling the gases and standardizing the eight features. The dataset was split into a training set and a test set, and hyperparameter tuning was performed to optimize the model's performance. The model's performance was evaluated using metrics like F1-score, recall, precision, and accuracy. The performance was tested on the test data, and the results were obtained from the confusion matrix and SHAP plots.

Hyperparameter tuning is the process of selecting the best combination of hyperparameters for a machine learning algorithm to achieve the best performance on a given dataset. Hyperparameters are parameters that are set before the machine learning algorithm is trained, such as learning rate, number of hidden layers, and number of neurons per layer.

Hyperparameter tuning involves selecting the values of these hyperparameters that result in the best performance of the machine learning algorithm. This is typically done by training the algorithm with different hyperparameter combinations and evaluating its performance on a validation dataset. The combination of hyperparameters that results in the best performance on the validation dataset is then selected as the optimal hyperparameters. This process is important to avoid overfitting and achieve the best possible accuracy in the model's predictions.

The F1-score is a measure of a binary classification test's accuracy. It is calculated as the harmonic mean of precision and recall, two other commonly used measures.

Precision measures the proportion of true positive results among the total predicted positive results, while recall measures the proportion of true positive results among the total actual positive results.

The F1-score balances both precision and recall, giving an overall measure of a test's accuracy. It ranges from 0 (worst) to 1 (best). A higher F1-score indicates a better balance between precision and recall.

Recall is a performance metric in machine learning that measures the ability of a model to correctly identify all relevant instances, also known as true positives (TP), from a dataset. It is calculated as the ratio of true positives to the sum of true positives and false negatives (FN):

Recall = TP / (TP + FN)

In other words, recall measures the proportion of actual positive instances that are correctly identified by the model. A high recall score indicates that the model is able to capture a large number of relevant instances in the dataset, while a low recall score indicates that the model is missing many relevant instances.

5th and 6th paragraph

The researchers used a machine learning technique called SHAP to figure out which features were most important for accurately predicting the different gas environments. They used SHAP to look at both the global importance of each feature (how much it contributed overall to the model's accuracy) and the local importance (how much it contributed to the model's accuracy for each individual data point). The SHAP plot shows which features were most important for accurate predictions, with the most important features listed at the top. The plot also shows which feature values were helpful (represented by red dots) or unhelpful (represented by blue dots) for accurate predictions.

Certainly! In this study, researchers used a technique called "disparity sensing" to identify different gases using graphene-based sensors. The graphene sheets were modified with different molecules, which caused changes in the electrical properties of the graphene when exposed to different gases. The researchers then used machine learning models to analyze the data and accurately identify the different gases. They

compared the performance of three different models (XGBoost, KNN, and Naive Bayes) and found that all of them had high accuracy in identifying the different gases.

To understand how the machine learning models were making their predictions, the researchers used a method called SHAP (SHapley Additive exPlanations). This allowed them to identify which features (i.e., the different electrical properties of the graphene) were most important in making accurate predictions, and how those features were related to each other. The researchers found that certain features were more important for certain gases, which suggests that different gases interact with the modified graphene in different ways. Overall, the study demonstrates the potential of using graphene-based sensors and machine learning to accurately identify different gases.

Disparity sensing is a technique that involves using the interaction between graphene and gas molecules to identify and differentiate between different gas environments. This technique relies on the concept of van der Waals bonding between graphene and gas molecules, which results in changes in the electronic properties of graphene. These changes can be measured using a field-effect transistor (FET) device, which can then be used to identify the specific gas molecules present in the environment. The disparity sensing technique can be combined with machine learning algorithms to achieve accurate identification and classification of different gas environments based on their unique electronic signatures.

7th paragraph

The SHAP values were used to analyze the importance of each feature in the developed models. The order of feature significance was different from the order of ANOVA p-values, indicating that p-values are not suitable for explaining feature contributions to the model. The SHAP values showed that lower values of some features were correlated with better gas identification performance, while higher values of other features were correlated with better performance. The scattering-based features generally proved to be more significant in determining the model performance.

ANOVA (Analysis of Variance) is a statistical method used to compare means of two or more groups to determine whether there are any significant differences between them. It assesses the variability within and between groups and tests the null hypothesis that the means of all groups are equal. If the p-value of the test is less than the chosen significance level (usually 0.05), then the null hypothesis is rejected and it is concluded that at least one group is significantly different from the others. ANOVA is widely used in scientific research to analyze experimental data and compare the effects of different treatments or interventions.

The p-value is a statistical measure that is used to determine the significance of the results obtained from a hypothesis test. In hypothesis testing, researchers propose a null hypothesis (H0) that assumes that there is no significant difference or relationship between two variables, and an alternative hypothesis (H1) that assumes that there is a significant difference or relationship between the two variables.

After conducting a statistical test, the p-value tells you the likelihood of obtaining the observed results (or more extreme results) assuming that the null hypothesis is true. If the p-value is very small (typically less than 0.05), it is generally accepted that the null hypothesis can be rejected and the alternative hypothesis is more likely to be true.

However, the passage you provided suggests that the p-values were not sufficient to explain the feature contributions to the model, meaning that the significance of the features could not be accurately determined based solely on their p-values. Instead, the authors used SHAP values to better understand the importance of each feature in the model.

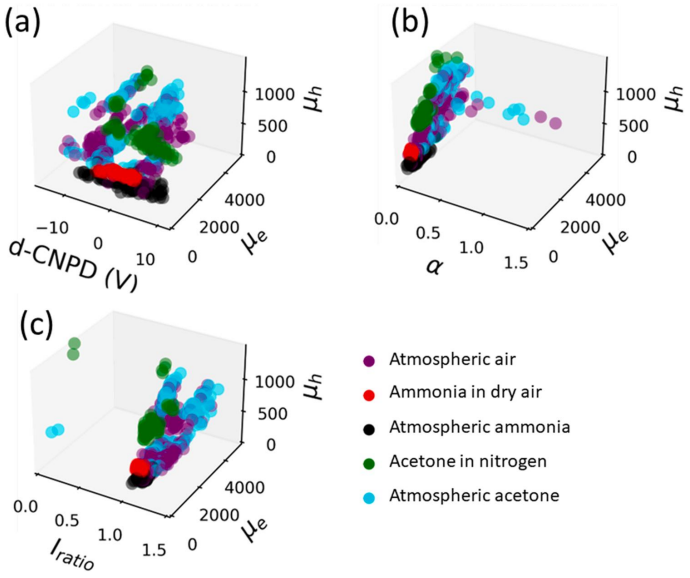
8th paragraph

The researchers tested the models they developed by including blank atmospheric air in the training and testing process. They found that the models had lower accuracy, precision, recall, and F1-score because it was difficult to differentiate between blank atmospheric air and atmospheric acetone. This is because the disparity measurements relied on electric field modulation, which is more sensitive to polar gas environments

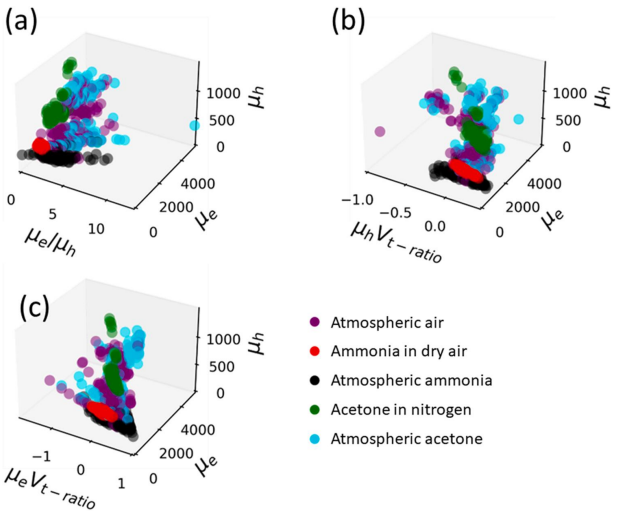
(e.g., atmospheric water/O2 electrochemical couple) than weakly polar gases (e.g., acetone).

They also tested the models using 7 features instead of 8 to determine if one feature, α, was not statistically significant as suggested by the ANOVA F-test. They found that the results were similar to the models trained with 8 features, confirming that α is not statistically significant and that the p-value is a good metric for feature selection, but not for determining the global feature importance.

The ANOVA (Analysis of Variance) F-test is a statistical test used to determine if there are significant differences between the means of two or more groups of data. It compares the variation between group means to the variation within groups. The F-statistic is calculated as the ratio of the variance between the groups to the variance within the groups. A high F-value indicates that the variation between groups is greater than the variation within groups, indicating that there are significant differences between the groups. The F-test is commonly used in hypothesis testing to determine if there is a significant difference between groups, and it can also be used for feature selection in machine learning.



3D plots of the various gas environments using three sets of the eight ML features, with two axes fixed to the most statistically significant features (μe, μh). a)
Plot of the d-CNPD vs μe, μh b) plot of α vs μe, μh, c) plot of Iratio vs μe, μh.

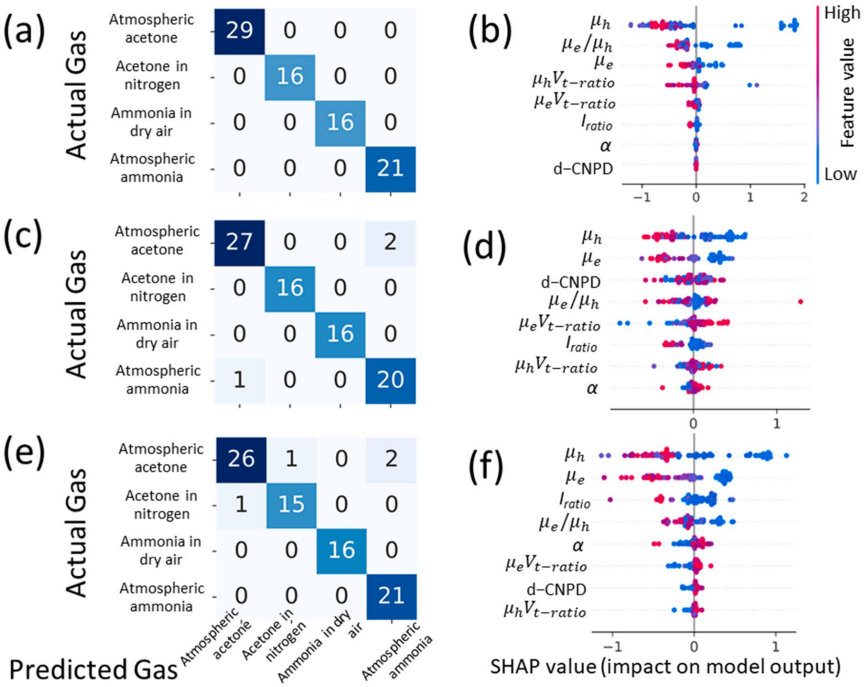


3D plots of the various gas environments using three sets of the extracted eight ML features with two axes fixed to the most statistically significant features
(μe, μh). a) Plot of μe/μh vs μe, μh. b) Plot of the change in hole mobility after Vt application relative to before Vt application (μhVt− ratio) vs μe, μh. c) Plot of the change
in electron mobility after Vt application relative to before Vt application (μeVt− ratio) vs μe, μh.

Conclusion

The researchers developed three models that can detect different gases in the air using an electronic nose made with graphene. The best performing model was able to correctly identify ammonia, acetone, and mixtures of these gases in the air with 100% accuracy. When blank atmospheric air (without any added gases) was included, the model still performed well but with slightly lower accuracy. The researchers used a method called SHAP to understand which features (or characteristics) of the electronic nose were most important for accurate detection of the gases. By studying how the graphene-molecule complex interacts with the gases and changes when an electrical voltage is applied, the researchers were able to create a highly accurate electronic nose for detecting gases in the air.

An electronic nose (e-nose) is a device that mimics the sense of smell of humans and animals by detecting and recognizing different odors or chemical compounds in the air. It consists of an array of chemical sensors that detect the presence of different gases or volatile organic compounds (VOCs) in the air and convert them into an electrical signal. The electrical signal is then processed and analyzed by a computer to recognize and identify the odor or chemical compound. E-noses have various applications in industries such as food and beverage, agriculture, environmental monitoring, and medical diagnosis.



Confusion matrix and SHAP plot of the test data. a) Confusion matrix for the XGBoost model, and b) the SHAP explanation of the feature contributions. c) Confusion matrix for the KNN model, and d) the SHAP explanation of the feature contributions. e) Confusion matrix for the Naïve Bayes model, and f) the SHAP explanation of the feature contributions.