

Gas Sensing Properties of Graphene-Rb-Based Sensor for Liquefied Petroleum Gas and Hydrogen

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ABSTRACT

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Rb-modified graphene powder was prepared by the chemical route synthesis method in this paper. X-ray powder diffraction (XRD) and scanning electron microscopy (SEM) were used to characterize the composition of the crystalline phase and the morphology of the prepared gas-sensitive materials, respectively. In particular, the study focused on the sensing behaviours of Graphene-Rb-based sensor towards power transformer fault gases such as hydrogen and liquefied petroleum gas. The molar concentration (M) level of Rb additive was varied systematically from 0.5 M to 2.0 M. The highest value of sensitivity factor (SF) of ~ 200 for 1.0M Rb-modified graphene to LPG gas was obtained at considerably lower temperature of 390K and for Hydrogen gas the highest value of sensitivity factor (SF) of ~ 225 for 1.0M Rb-modified graphene at considerably lower temperature of 380K. The selectivity value is found to be maximum ~ 16 for 1.5M of Rb-modified graphene for LPG and ~ 32 for 2.0M of Rb-modified graphene for Hydrogen gas.

Keywords : LPG Sensor; Hydrogen Sensor; Rb-modified Graphene; Sensitivity; Selectivity.

I. INTRODUCTION

Carbon the 6th element in the periodic tables has always remains a fascinating material to the researcher and technologist. Diamond, graphite, fullerenes, carbon nanotubes and newly discovered graphene are the most studied allotropes of the

carbon family. The significance of these material can be understood as the discovery of fullerene and graphene has been awarded noble prizes in the years 1996 and 2010 to Curl, Kroto & Smalley and Geim & Novalec, respectively. After the flood of publications on graphite intercalated [1], fullerenes (1985) [2], and carbon nanotubes (1991) [3], graphene have been the

subject of countless publications since 2004 [4, 5]. Graphene is a flat monolayer of carbon atoms tightly packed into a two-dimensional (2D) Honeycomb lattice; completely conjugated sp^2 hybridized planar structure and is a basic building block for graphitic materials of all other dimensionalities (Figure 1). It can be wrapped up into 0D fullerenes, rolled into 1D nanotube or stacked into 3D graphite.

In 2004, Geim and co-workers at Manchester University successfully identified single layers of graphene in a simple tabletop experiment and added a revolutionary discovery in the field of nano science and nanotechnology. Interest in graphene increased dramatically after Novoselov, Geim et al. reported on the unusual electronic properties of single layers of the graphite lattice. One of the most remarkable properties of graphene is that its charge carriers behave as massless relativistic particles or Dirac fermions, and under ambient conditions they can move with little scattering. This unique behavior has led to a number of exceptional.

Phenomena in graphene [4]. First, graphene is a zero-band gap 2D semiconductor with a tiny overlap between valence and conduction bands. Second, it exhibits a strong ambipolar electric field effect so that the charge carrier concentrations of up to 10^{13} cm^{-2} and room-temperature mobility of $10000 \text{ cm}^2\text{s}^{-1}$ are measured. Third, an unusual half-integer quantum Hall effect (QHE) for both electron and hole carriers in graphene has been observed by adjusting the chemical potential using the electric field effect [5, 6]. It has high thermal conductivity with a value of 5000 WmK^{-1} for a single-layer sheet at room temperature. In addition, graphene is highly transparent, with absorption of 2.3% towards visible light [7, 8]. Narrow ribbons of graphene with a thickness of 1-2 nm are, however, semiconductors with a distinct band gap and these can be used to produce transistors [9-11].

In last couples of years, graphene has been used as alternative carbon-based nanofiller in the preparation of polymer nanocomposites and have shown improved mechanical, thermal, and electrical properties [12-19]. The recent advances have shown that it can replace brittle and chemically unstable indium tin oxide in flexible displays and touch screens [20-21]. It is well established that the superior properties of graphene are associated with its single-layer. However, the fabrication of single-layer graphene is difficult at ambient temperature. If the sheets are not well separated from each other than graphene sheets with a high surface area tend to form irreversible agglomerates and restacks to form graphite through p-p stacking and Vander Waals interactions [22, 23]. Aggregation can be reduced by the attachment of other small molecules or polymers to the graphene sheets. The presence of hydrophilic or hydrophobic groups prevents aggregation of graphene sheets by strong polar-polar interactions or by their bulky size [24]. The attachment of functional groups to graphene also aids in dispersion in a hydrophilic or hydrophobic media, as well as in the organic polymer.

Therefore, an efficient approach to the production of surface-functionalized graphene sheets in large quantities has been a major focus of many researchers. The goal is to exploit the most frequently proposed applications of graphene in the areas of polymer nanocomposites, super-capacitor devices, drug delivery systems, solar cells, memory devices, transistor devices, biosensors and electromagnetic/microwave absorption shields. Before submitting your final paper, check that the format conforms to this template. Specifically, check the appearance of the title and author block, the appearance of section headings, document margins, column width, column spacing and other features.

II. METHODS AND MATERIAL

Rb-modified graphene is synthesized by chemical route method and the synthesized material was used for LPG and Hydrogen gas sensing application. To prepare Rb-modified graphene, rubidium chloride (RbCl), graphite (particle size 40 μm), conc. sulphuric acid (H_2SO_4 , 98%), sodium nitrate (NaNO_3), potassium permanganate (KMnO_4), and hydrogen peroxide (H_2O_2 , 30 wt. %) were obtained from S.D.fine Chem. Graphite, NaNO_3 , and KMnO_4 were used as obtained. Deionized water was used in the synthesis to ensure high purity of sample.

Structural and physiochemical properties of the resulting pellets were explored with the help of X-ray powder diffraction (XRD) and scanning electron microscopy (SEM) and gas sensing characteristics by four probe analysis. The Surface morphology of the synthesized products was observed using scanning electron microscopy (SEM) (JEOL, JSM-IT300). For the identification of crystal phase and to calculate various structural parameters of the obtained samples, X-ray diffraction (XRD) pattern were recorded using XPERT-PROMPD X-ray diffractometer, with $\text{CuK}\alpha$ radiation ($\lambda = 1.5405 \text{ \AA}$) in the 2θ range of 10-60. Gas sensing experiments were carried out in a sealed chamber. To measure conductivity, a Keithley 2400 source meter was used.

III. RESULTS AND DISCUSSION

To characterize the performance of the fabricated Graphene-Rb-Based Sensor device as a gas sensor, we measured the change in its resistance upon exposure to varying relative temperature of the sample inside the chamber with different molar concentration of Rb from 0.5M to 3.0M. Figure 1(a) and (b) shows variations of sensitivity with the temperature at different concentration levels of Rb-modified graphene for different molar concentration for LPG and H_2 gas (at 1000 ppm). It is clear that the SF is

maximum of ~ 200 for 1.0M Rb-modified graphene to LPG gas was obtained at considerably lower temperature of 390K and for Hydrogen gas the highest value of sensitivity factor (SF) of ~ 225 for 1.0M Rb-modified graphene at considerably lower temperature of 380K. The gas sensing response shows good dependence on the LPG and H_2 concentration as compared to other Rb concentration.

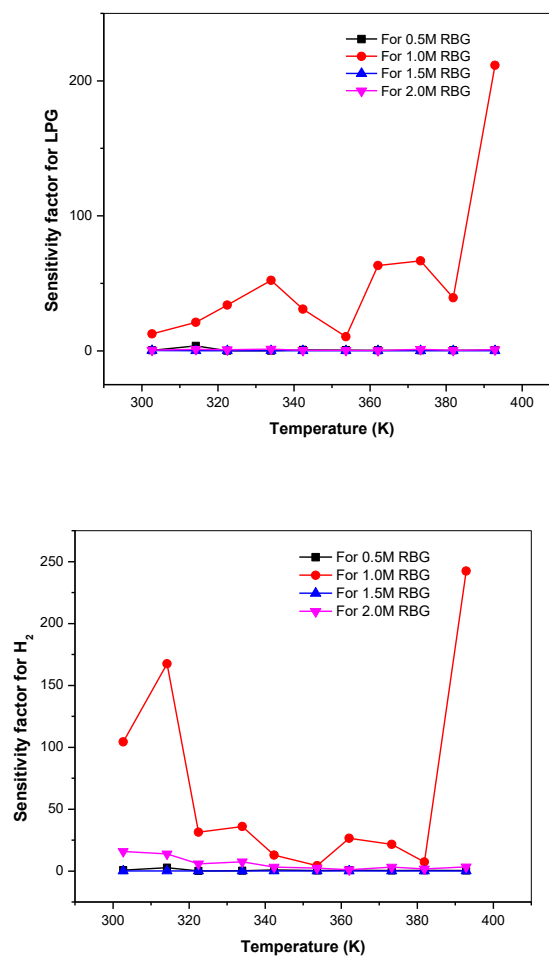


Figure 1 (a) variation SF with operating temperature for different molar concentration of Rb (at 1000 ppm of LPG).

(b) Variation SF with operating temperature for different molar concentration of Rb (at 1000 ppm of H_2).

The variation of selectivity with Rb molar concentration LPG and H_2 . Here, the selectivity is defined as the ratio of SF for test gas and interfering

gases (at the same concentration of 1000 ppm) at the optimized operating temperature for the test gas. The optimum concentrations of Rb-modified graphene as regards the selectivity for LPG are found to be maximum ~ 16 for 1.5M of Rb-modified graphene for LPG and ~ 32 for 2.0M of Rb-modified graphene for Hydrogen gas as shown in figure 2 (a) and (b).

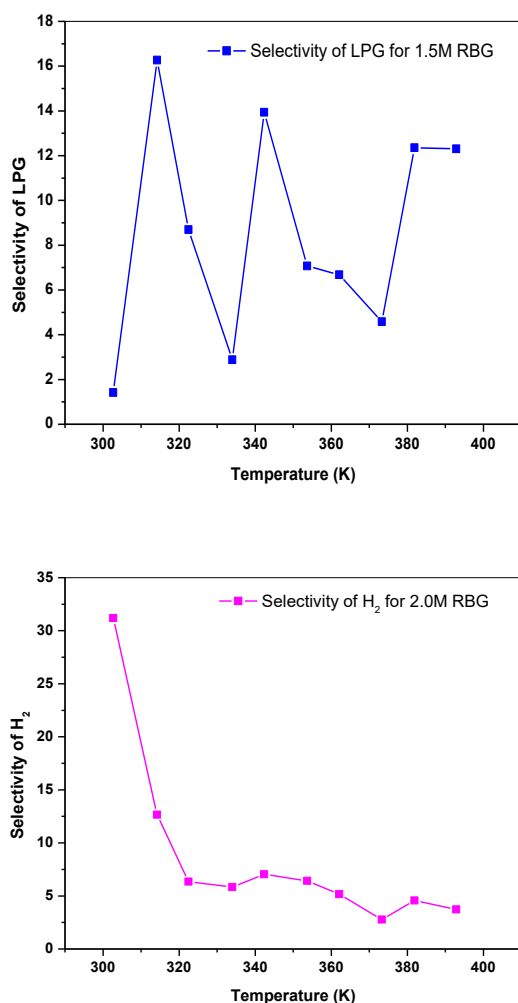


Figure 2 (a) Selectivity variation at different molar concentration of Rb (at 1000 ppm of LPG). (b) Selectivity variation at different molar concentration of Rb (at 1000 ppm of H₂).

The XRD pattern of pure graphene particles (Fig. 3) shows that the characteristic peak of Rb-modified graphene is at 26.6°. Figure 3 shows the XRD pattern of Graphene and Rb-modified graphene nanoparticles at different molar concentration of Rb. The peak position of all the XRD pattern is in good

agreement. The XRD studies were carried out to understand the crystallinity and phase of the samples, respectively. The XRD pattern reveals that the Graphene nanoparticles are more amorphous in nature. With doping of Rb, it shows crystalline nature.

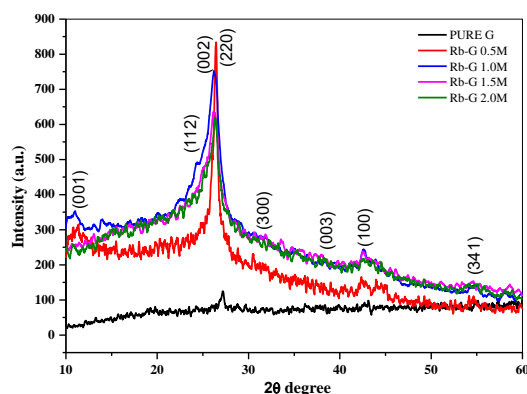


Figure 3. XRD pattern of Graphene and Rb-modified graphene nanoparticles at different molar concentration of Rb.

SEM images of the Graphene and Rb-modified graphene nanoparticles are shown in Figure 4(a), 4(b), 4(c), 4(d) and 4(e) for pure graphene and different molar concentration of Rb-doping such as 0.5M, 1.0M, 1.5M and 2.0M respectively. It is seen that the morphology of nanostructures strongly depends on the molar concentration of Rb. In Fig. 4 (a) the SEM image of highly oxidized graphite shows a smooth surface with tightly packed sheets, while the exfoliated sample exists as transparent, wrinkled sheets, indicating these layers are exfoliated to a very large extent. They are well organized. In the further SEM images 4(b), 4(c), 4(d) and 4(e) it is clearly seen that by doping of Rb the morphology changes from wrinkled sheets to granular like structure. Increasing molar concentration of Rb can cause further change in the morphology of samples.

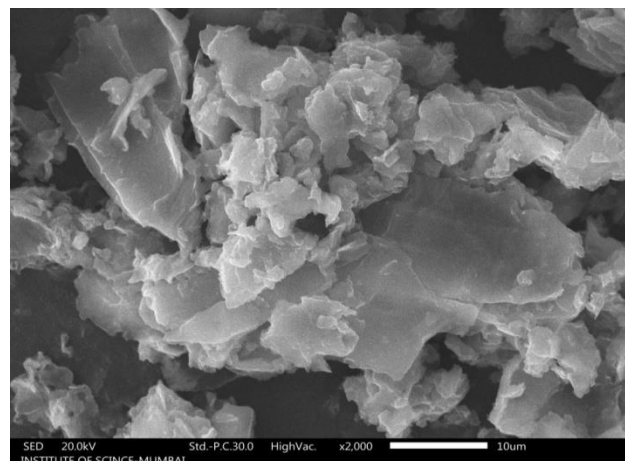
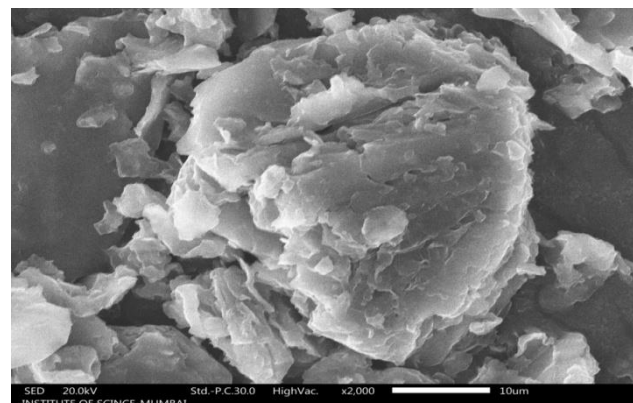
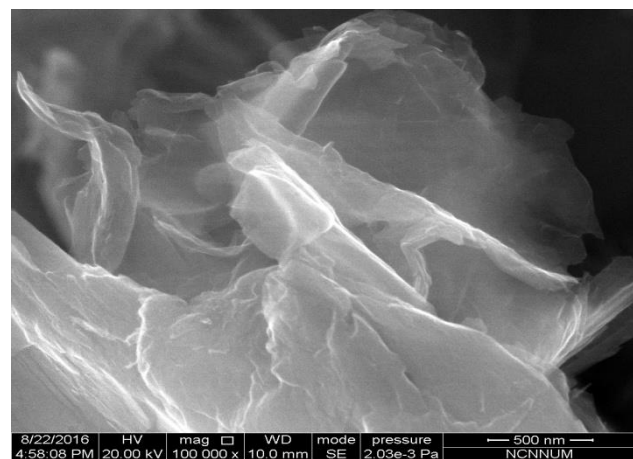
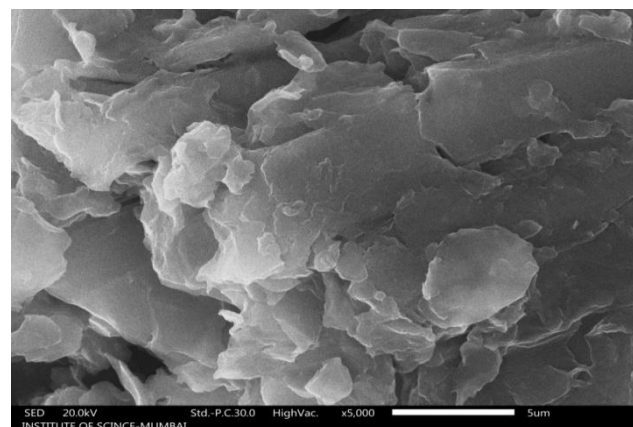
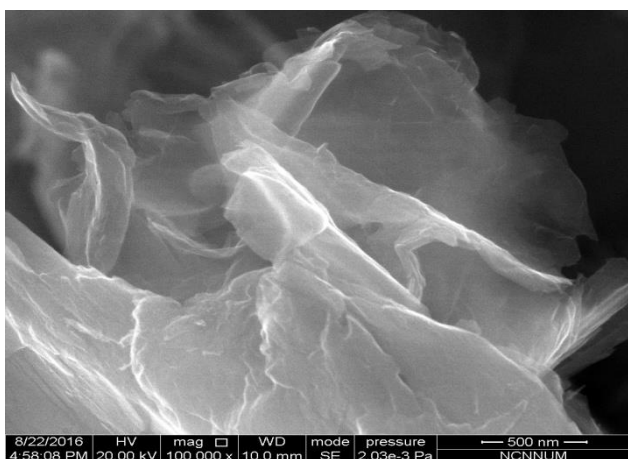


Figure 4. SEM images of the Graphene and Rb-modified graphene nanoparticles

IV. CONCLUSION

In the summary, we successfully demonstrated LPG and H₂ sensing application by Rb-modified graphene composite. The as-prepared sample shows good sensing and operating temperature response. The sample shows good selectivity characteristics. These high sensitivity and wider range characteristics are achieved by using considerably lower concentration level of Rb additive as compared to the reported data. The main achievement of present work is low operating temperature, which is much below auto-ignition temperature of LPG and H₂. The graphene sensor shows significant conductance changes when exposed to various concentrations of testing gasses in air. The response time of the sensor is less than 10 s, suggesting that the device shows fast response to LPG and H₂ gas. Because of the weak interaction between gases and graphene, the device response is rapid and reproducible. In the future, we will investigate the feasibility of using graphene sensors for environmental monitoring and carry out surface fictionalization of the graphene layers to enhance the selectivity of the sensor.

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