1.1 Thesis Abstract

The research communities have focused a great deal of attention on developing and advancing organic thin-film transistors (TFTs) for various electronic and sensing applications. These TFTs have many potential applications in the era of memory devices, organic LEDs, gas sensors, medical applications and many more. With the continuous research progress and swift developments in deposition methodologies, there has been a noticeable advancement in the existing solution-processable, low -voltage, low-cost wide area-based TFTs to fulfill various electronic and sensing applications. Moreover, the organic semiconductor-based thin film transistors have gained more interest due to their ability to use in flexible electronics, such as flexible LEDs, displays, keyboards, smartphones, sensors, memories, etc., and in flexible smart cards, such as RFID (Radio Frequency Identification) tags, etc. Exploiting a broad advantage of TFTs, this thesis broadly discusses the advancement in the fabrication and characterization of organic semiconductor-based TFTs and their utilization in enhanced ammonia sensing/detection.

This thesis contains six chapters, which have been organized simply and concisely to explore the advancement in the fabrication and characterization of the Organic TFTs (OTFTs) and their applications as selective ammonia gas sensor. Each chapter begins with a brief description of its contents, followed by motivation for the research, and further supported by graphical illustrations.

The thesis briefly discusses the evolution of TFTs, the basic charge transport phenomenon in organic semiconductors, various deposition techniques, challenges in the deposition of organic semiconducting channel, and gate oxide film of organic TFTs in ***Chapter 1***. This chapter also outlines various architectures, charge transport mechanism in TFTs,

application of OTFTs in gas sensing, literature survey/motivation behind OTFT-based ammonia sensors, and resources utilized in the device fabrication and sensing characterization/measurements.

Chapter 2, dealing with the gas-sensing applications of organic polymer nanocomposite-based thin-film transistor in ammonia sensing application. This chapter explores the introduction of organic TFT in gas sensing application, device fabrication, followed by characterization properties of the thin film, various sensing results, and sensing mechanisms incorporated in fabricated organic polymer nanocomposite-based TFT for ammonia sensor. It has been found that the introduction of the 2D MoS_2 flacks in the P3HT polymer matrix enhances the sensing response of the polymer nanocomposite matrix towards ammonia gas at room temperature operation (25 ºC), and is almost independent of relative humidity (RH) variation.

In ***Chapter 3***, a cost-efficient, minimal-wastage facile solution-processed floating film transfer method (FTM) has been discussed for the controlled morphological growth of organic semiconductor channel and further utilized for the gas sensing application. This chapter envisages a low-cost spin-deposited LaZrOx high-k dielectric layer for low-voltage OTFT device. The synthesized dielectric possesses a high band gap, low leakage current density, high stability, and the capability to generate a high saturated current even at low applied voltage. This chapter further explores various device characteristics and sensing results. It also investigates the device physics and sensing mechanism toward the target ammonia gas.

Chapter 4 includes novel high-k bilayer dielectric film and Au/P3HT nanocomposite organic semiconductor channel in the fabrication of organic TFT for ammonia sensing application. The introduction of TiO_2 and HfO_2 layers in the TiO_2/HfO_2 bilayer utilizes the advantages of both layers in terms of high dielectric constant (large capacitance per unit area) and high band gap (minimize leakage current density), which are further suitable for low voltage operation of the OTFT with the high saturated current. The bilayer dielectric also reduces the trap charge density at the dielectric/semiconductor interface and reduces the threshold voltage of the transistor. Moreover, introducing Au (gold) nanoparticles in the P3HT polymer matrix enhances the charge transport facility, the chain ordering, sensing response, and response/recovery time of the fabricated sensor towards ammonia gas.

Chapter 5 illustrates a novel high-k hybrid dielectric material as a gate oxide film and P3HT/g-C_3N_4 nanocomposite organic semiconductor channel for the fabrication of flexible organic TFT to obtain enhanced ammonia gas sensing performance. This work utilizes a hybrid ZrOx/PMMA/PMCF dielectric layer, which has the advantages of both the polymer dielectric (PMMA/PMCF) in terms of the smooth film (free form charge carrier scattering), flexible nature, etc., and the use of high-k inorganic dielectric (ZrOx) in terms of high dielectric constant (~ 22), etc. for the fabrication of low-voltage flexible organic TFT. In this chapter, the extensive study of the device and sensing characteristics of the fabricated flexible device has been thoroughly discussed. Moreover, the introduction of the 2D g-C_3N_4 in the pristine P3HT polymer matrix also demonstrates a better sensing characteristic due to improved π-π carriers' delocalization over the backbone of the polymer matrix.

Chapter 6 presents a summary of the outcomes of the study undertaken during the work. The overall conclusion drawn from the study has been enunciated. The study revealed that it is possible to bring down the operating voltage of conventional OTFTs by using suitable high-k dielectric material as a gate oxide layer. It can also be concluded that with the suitable selection of organic polymers/nanocomposite materials and hybrid dielectric (Polymer/Inorganic) materials with certain optimization and deposition techniques, one can fabricate a cost-efficient, flexible organic TFT device in the area of electronic and sensing applications. This chapter further outlines the future scope of this extensive study.

1.2 Evolution of Thin Film Transistors

Thin film transistors are a special class of metal oxide semiconductor field effect transistors (MOSFETs), where the transistor is very thin as compared to the plane of the device. It can be fabricated by coating/growth of an active semiconductor layer, dielectric/oxide layer, and metallic contacts over any kind of rigid substrate. The substrate type includes silicon, glass, flexible PET (Polyethylene Terephthalate), etc., for device fabrication. Although the concept of the field effect transistor's idea was first patented by Julius Edger Lilienfeld in 1925 and in 1934 by Osker Heil, it began to receive significant attention over the late 1970s. The rising competition in the field of MOSFET, which uses the complex and costly fabrication process on a silicon substrate, motivated researchers to find an alternative to conventional MOSFETs. In addition to that, the necessity of low-cost portable electronics for large-area applications, such as in memories, displays, sensors, etc., motivated scientists to develop a low-cost, simple TFT device in the early 1970s [1]. The three scientists, Spear, Ghaith, and LeComber, described TFT in 1979 using hydrogenated amorphous silicon (a-Si: H) as a semiconductor material. After that, several active and dielectric layer modifications were done to obtain a reliable, high-performance TFTs device. In the 1980s, silicon-based TFTs gained a lot of attraction due to their large potential application in LCDs (liquid crystal displays) and proved itself as a very important device in the area of emerging electronics. Later, in the 1990s, TFTs with organic semiconductors as active layers were introduced with high electron mobility