Recent Progress on Titanium Dioxide-Based Humidity Sensor: Structural Modification, Doping, and Composite Approach

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Abstract Advancement to the performance of humidity sensor is an importance subject due to its diverse role in multitude of applications. The trend shows that researchers are using various approaches to tackle the performance issues of humidity sensor. This paper reviews recent progress on the improvement of titanium dioxide-based humidity sensor performance through structural modification, doping and composite effect. These three approaches have seen tremendous success in elevating the humidity sensing performance of titanium dioxide-based humidity sensor in term of higher sensitivity, shorter response/recovery time, and higher stability. The main advantages of the modified nanostructures are that they offer huge surface area which is beneficial for the adsorption of water molecules and directional pathway

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for the flow of charge carrier. Meanwhile, doping and formation of composite have contributed by having a positive synergistic effect in term of hydrophilicity, electrical conductivity, surface reactivity etc. In this work, efforts to improve the humidity sensing performance by various researchers working on this material are presented and discussed.

Keywords Humidity sensor · Titanium dioxide · Nanostructure · Doping

1 Introduction

The importance of metal oxide semiconductors is growing with each passing year. Materials such as ZnO, NiO, SnO, MgO etc. are constantly at the centre of research subject from researcher from all over the world. This is due to the advantageous properties of metal oxides namely their non-toxicity, high chemical stability, intrinsic high sensing characteristic, low cost, and their abundance in nature. These compounds are formed when the electron from metal element is released to the oxygen atom resulting in oxides with different bonds and electronic structures [1].

One such metal oxide is titanium dioxide (TiO_2) or titania. TiO_2 is semiconductor with bandgap energy of 3.0–3.2 eV [2]. Since the discovery of its photocatalytic capability in 1972 [3], interest in this compound continue to rise. Furthermore, in 1988, Gratzel et al. [4] have introduced a new type of solar cell which utilizes TiO_2 as the photoanode in dye-sensitized solar cell. Nowadays, TiO_2 have found its way in various type of application such as pollutant degradation photocatalyst [5], photochemical water splitting, UV photosensor [6], biosensors [7], and chemical sensors [8].

One of the important chemical sensors is the humidity sensor. It is a device which can detect the level of water molecules in the air. It is used in wide variety of fields such as home appliances, agricultural, weather forecasting, and health monitoring. In the light of recent advancement in Internet of Things (IoT) and fourth industrial revolution (IR 4.0), the trend of application is shifting toward interconnected sensors and wearable devices. Depending on the transduction mechanism, humidity sensor can be divided into several types namely resistive, capacitive, and optical [9]. Humidity is often expressed in term of relative humidity (RH). It can be defined as the ratio of the amount of water vapour present in the atmosphere to the maximum amount that the atmosphere can hold [10]. RH is usually stated in unit of percentage.

Experimental works and analyses are being carried out by numerous groups with the aim to promote the performance of TiO_2-based humidity sensor mainly in term of sensitivity, response/recovery time, hysteresis, linearity and stability with various degree of success. However, a comprehensive review on this subject remained scarce. Herein, recent progress on the TiO_2-based humidity sensor is presented and discussed. Emphasis is placed on efforts which were based on structural modification, doping process and composite formation with other materials.

2 Effect of Different Types of Nanostructures

It is widely considered that humidity sensing mechanism are largely influenced by the structural or the shape of the material [11]. Herein, various kind of structures and their effects to the performance of humidity sensor are discussed.

2.1 TiO_2 1-D Nanostructure

1-D nanostructured material refers to a material with one of its dimensions is larger than the other two dimensions. These materials usually are associated a very high aspect ratio. Examples of these type of structure includes nanorods [12], nanowires, nanotubes, and nanofibers. This type of structure offers good carrier transport along its structural axis. The direct pathway of the movement of electron or hole enable faster response with high sensitivity.

Jyothilal et al. [13] have reported on the fabrication of humidity sensor using slanted TiO_2 nanorods produced using electron beam evaporation. They have achieved high sensitivity with fast response and recovery time of 145 and 120 ms respectively. It is suggested that the morphology has helped increase the carrier concentration leading to improved protonic conduction. Their sensor also showed promising potential as breath analyzer.

TiO_2 nanotube structure have been tested by Farahani et al. [14] as the humidity sensor. They have claimed that the structure is super-hydrophilic, allowing full absorption of water molecules. Their sensor recorded responsivity of 300%.

Meanwhile, Li et al. [15] have experimented with humidity sensor made of TiO_2 nanowires. Sensitivity of 280 pF/% RH was recorded at low RH (7% RH to 33% RH). It is supposed that the cross-linked nanowire structure improves charge conduction leading to the increase in sensitivity at low RH environment.

2.2 TiO_2 2-D Nanostructure

2-D nanostructures are made of with thin layer structure. Nanosheet, nanowall, nanobelt, nanodisk, and nanoflakes are some of the examples of 2-D nanostructure. These types of structures were also being utilized to increase the performance of humidity sensor. For example, 2-D graphene [16, 17], NiO [18], WS_2 [19] and MoS_2 [20] have shown promising result as humidity sensor.

However, the use of TiO_2 2-D structure for the application of humidity sensor remained scarce. One report on TiO_2 nanosheet-based ultrasensitive humidity sensor have been published by Gong et al. [21]. The ultrathin petal-like structure shortened transporting paths and increase contact area between sensing material and electrodes. The self-assembled TiO_2 nanosheet yielded a nanoporous structure with high specific surface area. It is also argued that the ultrathin structure is rich with oxygen vacancy defect which is beneficial for humidity absorption. Up to 10 times improvement in sensitivity value was observed from the fabricated device.

2.3 TiO_2 3-D Nanostructure

3-D nanostructures usually encompassed three categories which are 3-D spatial ensembles of 1-D or 2-D nanostructure, 3-D nanoporous structures and 3-D hierarchical nanostructured materials [22]. Examples of this type of structure are nanoflower [23], nanoball, nanocube and interconnected pores. They are usually credited as having the largest surface area. Zhang et al. [24] reported on the quartz crystal microbalance (QCM) type humidity sensor using hollow ball, nanosphere and nanoflower TiO_2. The hollow ball structure which has the largest surface area showed the highest sensitivity of 33.8 Hz/%RH.

Similar nanoflower structure was also reported by Jeong et al. [25] who have fabricated resistive type humidity sensor with sensitivity of 485.7 $RH\%^{-1}$. Urchin-like structure have been reported by Wang et al. [26]. Figure 1 shows the examples of different types of TiO_2 nanostructures, and Table 1 gives a summary of TiO_2 nanostructure-based humidity sensor.

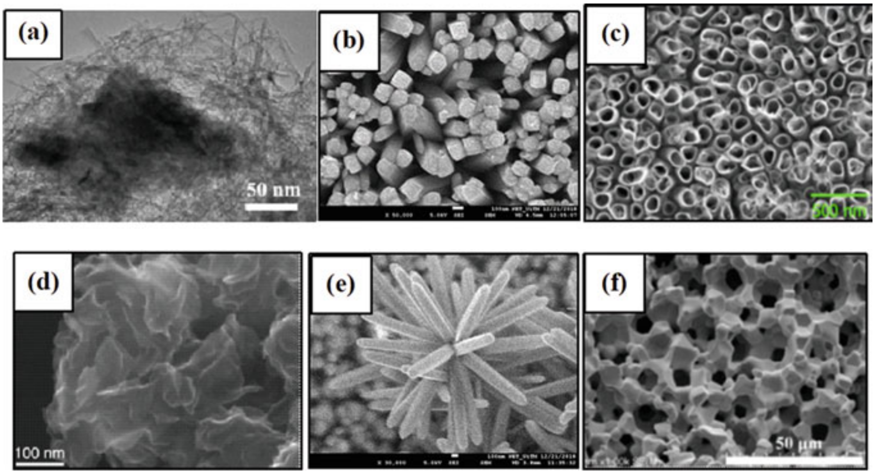

Fig. 1 Examples of different types of TiO_2 nanostructures: **a** TiO_2 nanowire, **b** TiO_2 nanorod, **c** TiO_2 nanotubes, **d** TiO_2 nanosheet, **e** TiO_2 nanoflower, **f** TiO_2 interconnected pores. Reproduced with permission from [15, 21, 27, 28]

Table 1 Summary of TiO_2 nanostructure-based humidity sensor

Structure type	Structure	Preparation method	Sensor type	Sensor performance	References
1-D	Nanorods	Electron beam evaporation	Resistive	Response time 145 ms Recovery time 210 ms	[13]
	Nanotubes	Anodization	Voltage	Sensitivity 300	[14]
	Nanowires	Alkali oxidation	Impedance	Sensitivity 280 pF/% RH	[15]
2-D	Nanosheet	Surfactant self-assembly	Impedance	Sensitivity 4-order of magnitude	[21]
3-D	Interconnected pores	Emulsion template	Impedance	Sensitivity 4-order of magnitude	[28]
	Nanoflower	Solution immersion	Resistive	Sensitivity 196	[29]
	Nanoflower	Hydrothermal and gravure printing	Resistive	Sensitivity 485.7 $RH\%^{-1}$	[25]

3 Effect of Doping

Doping is the act of adding specific amount of impurity into a host material in order to improve its properties. In the case of TiO_2, the doping could be divided into metal and non-metal doping. Dopant properties such as atomic radius and electronegativity would influence the effectiveness of a doping process [30].

3.1 Metal Doping

Li et al. [31] reported on the doping of niobium (Nb) into TiO_2. The pentavalent Nb^{5+} ion is a donor and will donate electron based on the following equation:

$$Nb_2O_5 \xrightarrow{2TiO_2} 2Nb_{Ti}^{\cdot} + 4O_o + 1/2O_2 + 2e'$$

This type of reaction would increase the electron concentration of TiO_2. Since electron conduction is responsible for the humidity detection mechanism, this has resulted in improved sensitivity of up to 13,705 pF/%RH in a capacitive-type humidity sensor.

Quartz crystal microbalance (QCM)-type humidity sensor made of Ru-doped TiO_2 nanofiber have been reported by Farzaneh et al. [32]. Using density functional

Table 2 Summary of doping effect on TiO_2-based humidity sensor

Dopant type	Dopant	Sensor type	Sensor performance	References
Metal	Nb	Capacitive	13,705 pF/%RH	[31]
	Ru	QCM	Four times more sensitive than undoped TiO_2	[32]
	Co	Resistive	Sensitivity five orders of magnitude	[33]
Non-metal	N	Resistive	Sensitivity five orders of magnitude	[34]
	GO	Optical	0.47 dB/%RH	[35]

theory (DFT) calculation, they have managed to show that the Ru-doped TiO_2 have significantly larger surface reactivity compared to undoped TiO_2. This have helped increase the water absorption ability, leading to the increase in sensitivity.

Meanwhile, Li et al. [33] presented Co as the dopant for TiO_2 for the application of resistive-type humidity sensor. Substitution of Ti^{4+} with Co^{2+} creates many defect sites and reduces the grain size, effectively increasing the humidity sensing performance.

3.2 Non-metal Doping

Humidity sensing properties could also be improved by doping with non-metal elements. Li et. al. [34] has reported on the N doping of ordered mesoporous TiO_2. Through XPS analysis, the resulting material showed the presence of Ti^{3+} defect sites. The defect sites would react with water molecules, releasing additional electron to the structure which would improve the sensitivity.

The sensitivity of optical-type humidity sensor could also be increased through non-metal doping as demonstrated by Ghadiry et al. [35] who added graphene oxide (GO) in TiO_2 solution which were then drop-casted on waveguides. GO helps in the diffusion of water molecules which bring significant changes to the refractive index. Table 2 shows the summary of doping effect on TiO_2-based humidity sensor.

4 Effect of Composite

Another approach to improve the performance of humidity sensor is through the formation of TiO_2 composite. Since it is quite challenging to simultaneously produce all the desired properties in a single material, synthesis of composite material has been gaining considerable attention [36]. Among the composite types which are commonly used are layered, core–shell, random mixture, and decoration structure. Composite materials are interesting as they often exhibit improved physical and chemical properties compare to each of the material alone.

Table 3 Summary of TiO_2 composite-based humidity sensor

Composite	Sensor type	Sensor performance	References
TiO_2/ZnO	Impedance	Sensitivity 0.4 orders of magnitude	[37]
TiO_2/VOPcPhO	Capacitive	Better sensitivity, lower hysteresis and reduced absolute threshold value	[41]
TiO_2/PVDF	Capacitive	Response time 45 s, recovery time 11 s	[39]
TiO_2/PLA	Capacitive	Response time 40 s, recovery time 20 s	[42]
$CoTiO_3/TiO_2$	Resistive	Sensitivity 157.23	[11]
$TiO_2/NaNbO_3$	Resistive	Sensitivity 125,512	[36]

Araujo et al. [37] have reported on the TiO_2/ZnO composite-based humidity sensor. Their result showed improved performance in term of increased sensitivity. Both metal oxides complimented each other properties. TiO_2 compensated the low hydrophilicity of ZnO while ZnO nanostructure improve the overall surface area of the sensor. TiO_2 naturally have high hydrophilicity because of the presence of Ti^{3+}/Ti^{4+} interface sites which is associated with dissociative transfer of water molecules [38].

Polymer is another material capable of forming beneficial composite with TiO_2. Mallick et al. [39] formed TiO_2 composite with polyvinylidene fluoride (PVDF) for the application of humidity sensor. They reported that the addition of TiO_2 increases the hydrophilicity of the sensor. Moisture absorption and settling were reported to increase at a more hydrophilic surface, leading to increase in the sensitivity of capacitive humidity sensors [40].

Some researcher worked with TiO_2 and organic-inorganic hybrid nano-composites, VOPcPhO [41]. Formation of this composite helped produced uniform voids across the surface of the material which are favourable for the absorption of moisture content. These voids also increase the surface porosity as evidenced through atomic force microscopy (AFM) result.

Adding a p-type semiconductor to n-type TiO_2 could form heterojunction structure as shown by Lu et al. [11]. Carrier transportation could be increased significantly through the decrease of electron current path at the heterogenous interface. Si et al. [36] reported a similar result when using $TiO_2/NaNbO_3$ nanocomposite. In their work, detection of humidity was achieved through the reduction of potential barrier height of the heterojunction due to the transfer of electrons from $NaNbO_3$ to TiO_2 upon exposure to moisture. Table 3 shows the summary of TiO_2 composite-based humidity sensor.

5 Conclusion

A review on the recent progress of the development of TiO_2-based humidity sensor have been presented. Engineering the shape of TiO_2 nanostructures into 1-D, 2-D and 3-D could bring advantageous properties to the performance of humidity sensor. In addition, doping process and formation of composite have also been shown to contribute to the increased performance of humidity sensor. Clearly TiO_2 have a very good potential to become a very efficient humidity sensing material thanks to the commendable effort by researcher on this subject. However, there are still gaps to be explored to further improve the performance of the device.

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