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Composite Effect on Zinc Oxide based Resistive Type Humidity Sensor Performance: A Preliminary Assessment

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Abstract— **Humidity sensors generally are capable of sensing and measuring substantial changes in water content in the air and converting them into an operational and functional signal in accordance with a specific rule of law. Single structure of pristine ZnO suffers in several pitfalls that severely limit the development of zinc oxide as humidity sensors, leading to unfavorable sensor performance. On the other hand, composites demonstrate elevated effects between closely packed surfaces, outperforming single structures. Surface modification effects emerge as a result of the intensification of nanostructures with active surface sensing sites, resulting in an improvement of surface-related properties. The integration of metal oxides with other materials to generate composite structure demonstrate an enhancement in sensitivity and/or other vital humidity sensing properties even further. In this review, various morphologies and composite structures have been presented, each with exceptional humidity sensing characteristics throughout a wide range of relative humidity levels. Apart from that, working principle and conduction mechanism of resistive type humidity sensor will be discussed and presented. From this review, it can be concluded that the effect of compositing ZnO with other materials contributed to the enhancement and improvement performance of the humidity sensor towards humidity sensing.**

Index Terms— **Zinc Oxide, Resistive Type Humidity Sensor, Composite Structure.**

I. INTRODUCTION

HUMIDITY, or the concentration of water vapor in the atmosphere, has a significant impact on ecosystems. High-accuracy relative humidity monitoring is an inevitable task and essential in several areas, including the environmental monitoring, foodservice industry, clinical practice, agriculture and plantation, industrial manufacturing, semiconductor foundry and biotechnology [1]–[4].

As a consequence, efforts should be directed on developing humidity sensors with fast response and recovery times, excellent reproducibility and stability, low hysteresis variation, high sensitivity, and the flexibility to operate at a variety of temperatures. A humidity sensing device based on resistive type humidity sensing principle drawn more interest due to thermal stability, excellent optical and electrical characteristic, better sensitivity, inexpensive and decent linearity [5].

Typical humidity sensors are constructed from a variety of materials, with metal oxide semiconductors being one of the most prominent. Due to its substantial ratio of surface-to-volume, minimal cost, superior sensitivity, simplicity in fabrication, fast response with improved sensitivity, and decent thermal and chemical stability, two dimensional metal oxide semiconductor sensing materials are commonly used as sensing materials [6], [7]. Moreover, due to their wider band gaps and greater surface areas, metal oxide semiconductor nanostructures exhibit unique physical, chemical, and optical characteristics that suitable for usage as sensing materials. The nanostructures high surface area has a substantial impact on the properties of the nanomaterials. With these excellent features, metal oxide semiconductor nanostructures are useful for a broad selection of applications. Among metal oxide semiconductor, zinc oxide (ZnO) has attracted fascinated consideration as a sensing material due to its availability and tunable surface structure, wide resistivity range, excellent chemical and thermal stability [8].

ZnO has appeared as a notable substance in the areas of sensing transducer and the fabrication of transparent conducting electrodes due to its stable n-type semiconducting material properties with large exciton binding energy and direct band gap, as well as distinctive electrical and optical properties [9]–[11]. Along with its structural merit such as morphological diversity, high crystallinity, and surface porosity, combine with advantageous such large specific surface area, high electron mobility, outstanding environment compatibility, low production cost, excellent thermal stability, and oxidation resistibility, zinc oxide (ZnO) is a suitable solution for

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numerous functional devices, including humidity sensors [5], [6]. However, the sensitivity towards humidity become an issue when detecting humidity with pristine metal oxide semiconducting material. Apart from that, other drawbacks of single-phase metal oxide-based humidity sensors are typically low response, longer response/recovery times, deficient linearity, and poor hysteresis [12]. To alleviate the issues related to sensing performance, the sensor might incorporate surface modification, doping, or the introduction of multi-compositional nanostructures.

An excellent method that has recently received a lot of attention is to couple two distinct semiconductors together, resulting in the production of a composite. As a consequence, a composite metal oxide semiconducting nanostructure should potentially demonstrate to be an efficient solution for improving sensor related performance parameters. When examined under identical experimental configurations, composite metal oxide sensors demonstrated substantially better sensing performance as such higher sensitivity and fast response than conventional sensors that composed primarily with one material [13]–[15].

The idea behind composite is coupling two types of semiconductor materials that owns dissimilar energy band gaps which allowed recombination of electron–hole pairs that directly modified material properties and thus by making composites of it, the excellent sensor performance is achievable [16], [17]. Moreover, metal oxide composite nanostructures are convincing candidates for humidity sensing applications because they are chemically and thermally stable, have a relatively high surface ratio for interaction with the humid environment than bulk thin film materials, high conductivity and, as an outcome, exhibit high sensitivity towards humidity [18].

Therefore, to construct a nanocomposite structure, one of the materials must be comprised of nanoscale materials. There are several types of nanocomposites, including mixed compound nanocomposite, bi-layer and multiple-layer films, decorative nanostructures with auxiliary nanoparticle elements, mixed nanostructures-nanostructure arrangement, core-shell structure, and branch-like structure as depicted in Fig. 1 [19]. Despite numerous publications on humidity-sensing materials and fabrication methods, hardly a few review articles have been published on the effect of composite structure on resistive type humidity sensor sensing performance. Thus, in this review, we addressed several significant works on zinc oxide humidity sensor that utilizing composite nanostructured as sensing materials and their improvements over pristine element.

II. MECHANISM OF RESISTIVE TYPE HUMIDITY SENSOR

Humidity sensors based on resistive or impedance type detect ambient in vapor counts, which significantly influence the electrical resistance or impedance of the active detecting layer [20]–[22]. For comparison purposes, increased humidity levels will frequently cause the electrical conductivity of the active sensing layer to increase while concurrently decreasing the systems resistance. The variations of resistance (or impedance) result from the response to substantial condensation of water vapors in an increased humid ecosystem, which occurs on the surface of sensing films [23]. The ionic functional hydroxyl groups of water molecules dissociate as a result of this phenomenon. The emergence of a mobile pathway for dissociated charges is then stimulated because of an increasing amount of condensed vapors and thus, helps facilitate an efficient conduction path for the hygroscopic layer. Eventually, as conductivity transpires, the system's electrical resistivity decreases [24].

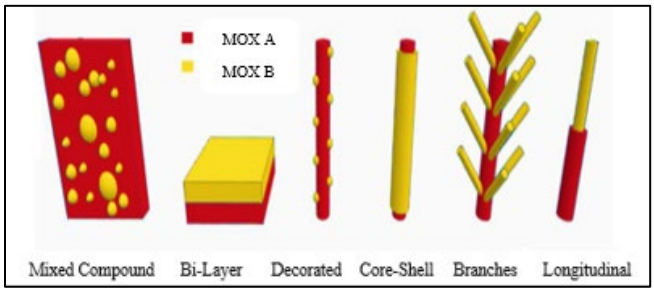

Fig. 1. Various types of metal oxide (MOX) composite structure according to their morphology [19].

Recently, Relative Humidity (RH) class humidity sensors are currently available on the market or in development in laboratory studies, and they are divided into three categories of sensing material which semiconductor type, organic polymer-based sensors, and organic/inorganic hybrid sensors [25]. All three types of hygrometric sensors employ changes in physical and electrical properties of the sensitive elements once exposed to a variety ambient humidity condition throughout the surrounding environment to provide a humidity quantification due to a certain amount of adsorption and desorption of water vapour molecules. It is worth to mention, that porous films are more sensitive to humidity than nonporous films [26]. Apart from that, the existence of intragranular porosity, as well as the distribution of pore size, are also important features in humidity sensors [27]. Therefore, in general, humidity is measured in hygrometer sensors by measuring either the electrical resistance (or impedance) of the sensitive material, which is proportional to the difference in the physics of any organic or inorganic synthetic material.

For resistive/impedance type humidity sensor, the conduction mechanism of the sensor can be subcategorized as ionic-conduction or electronic-conduction [28], [29]. Both electronic and ionic type humidity sensor estimate the moisture value by measuring the changes in conductivity of the sensor films as a proportion different of humidity levels. The operational approach of all semiconductor-based humidity sensors, either inherit ionic conduction, or electronic

conduction, is premised on superficial water layer adsorption associated with chemical adsorption, physical adsorption, and capillary condensation processes [30]. In electronic type mechanism, electrons are donated by the water molecules. These water molecules undergo chemical adsorption, enabling the electrical conductivity to improve or diminish. However, if the humidity sensor inherit ionic conduction mechanism, the sensor resistance (or impedance) decreases as the relative humidity value increases owing to the generation of a physical adsorption layer and condensation of water molecules in the capillary on the materials surface [31].

At the lower humidity environment, the active sensing layer adsorbs oxygen from the surroundings, trapping free electrons and resulting in the formation of adsorbed oxygen anions (O_2^-). Thus, due to this electron trapping, the concentration of free carriers in the sensing material films decreased significantly [32]. The negatively charged oxygen is electrostatically bonded to the sensor material positively charged metal ions, and the initial monolayer is chemisorbed as a result of the formation of a chemical interaction between the metal cation and the hydroxyl anion of the water under the influence of a substantial electrostatic field [33]. As the sensing material come in contact with water vapor and the humidity slightly increases, the chemical adsorption (chemisorption) process begins, and water molecules chemisorb on the sensing material surfaces accessible sites. During the chemisorption process, water vapor condensate onto the active surface of sensing material in response to the chemical reaction that occurs between the surface of the porous film, which is accompanied by the appearance of free-moving hydroxyl ions (OH^-). These dissociated hydroxyl ions (OH^-) commute further before being bound to the neighboring metal cations [34]. The chemically absorb water molecules released the trapped electron back to the sensing film. Thus, the resistance of the sensing element starts to reduce (conduction increased) as the relative humidity rises. Therefore, the early conducting stage can be credited to the electronic conduction that results from electron diffusion as depicted in Fig 2.

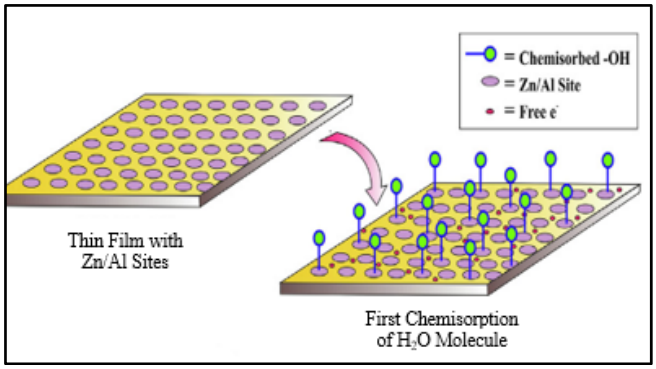

Fig. 2. Mechanism of electronic conduction at early conducting stage [7] .

As the humidity level elevated, the condensed vapors that continuously escalate compensate the stacked aquatic layers. The dissociative chemisorption process first forms the chemisorbed layer, then followed by the formation of the first physical adsorption (physisorbed) layer [35].

The formation of physically hydroxyl multilayers is currently in progress and compared to its chemically formed counterpart (chemisorption layer), the secondary layer is much less ordered. Following that, the approaching water molecules continue to intensify the development of the stationary layer known as the physisorbed layer, which now permeates the active surface. Due to the formation of double hydrogen bonds with the preformed chemisorbed layer, this secondary film has a rigid structure. However, because of the limitation imposed by the two hydrogen bonds, the top water molecule condensed cannot move freely. Therefore, the first physically-adsorbed layer is stationary, and no hydrogen bonds are formed between the water molecules in this layer, thus, no proton could be conducted at this point [36]. As a consequence, the predominant electrical conduction of the active layer can be attributed to electron tunnelling (trapped electron released back to film by the chemisorption water molecules), which occurs mostly between donor water sites.

As the surrounding humidity further rises, the thick layer of condensed water molecules on top of the sensing material facilitates proton conduction within the sensing membrane. In the secondary stage, the proton is the primary contributor of charge carrier, which is essential for ionic conductivity. These hygroscopic interactions on the active sensing layer assist in the dissociation of condensed water vapor into their respective ionic elements. Throughout this phase, migrating protons in the form of hydrogen ions (H^+) significantly contribute to electrical conductivity and diffusion of the proton occurs most prominently between adjacent hydroxyl ion (OH^-) carriers. As result, the freely-moving H^+ components predominantly leap among the arriving vapor, producing hydronium ions, H_3O^+, as by-products [37], [38]. Continuously, the hydronium carrier ions hydrated and decomposed into hydrogen cation and water, where each water molecule will have a slightly linked with hydroxyl group. The hydrogen cation from the hydronium ion continuously leaps from one water molecule to the next, which became source of electrical conduction. This tunnelling activity of the hydrogen cations is because of the excellent mobility of the proton in water vapor. In the theory of liquid decomposition by electrical currents, this mechanism of ionic conduction is known as Grotthuss chain reaction as shown in Fig.3 [39].

Therefore, at higher humidity levels, continuous ionic conduction due to proton tunnelling is responsible for becoming the dominant conducting mechanism, where sustained physisorbed water layers contribute to the enhanced mobility of hydrogen cation carriers as described through the Grotthuss mechanism. The emergence of carriers increases the capacitance of the sensors (due to increase of dielectric) while decreasing their resistance. Nevertheless, at relatively much

higher humidity environment, the water molecules condensed in the pore channels between grain boundaries or capillaries of the porous structure sensing material. This condensation of water molecules allowed the higher maneuverability of conducting ions freely across and between sensing membrane nanostructures and hence enhance the electrical conduction of the sensors [32], [40]. This conduction mechanism is dubbed as capillary condensation as illustrated in Fig 4.

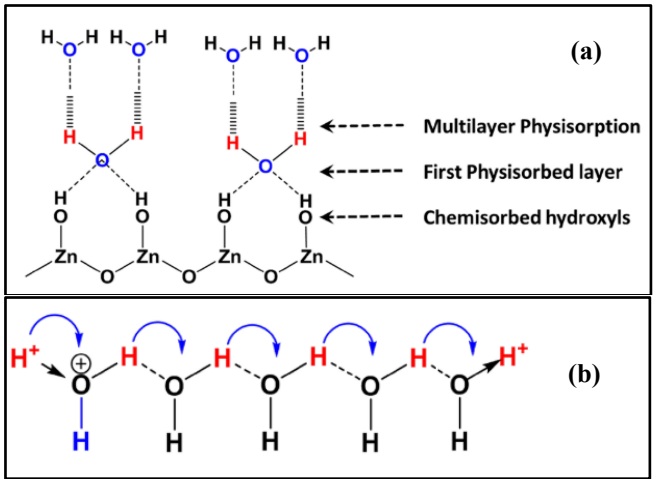

Fig. 3. Mechanism of ionic conduction as humidity level further elevated. (a) Water molecules absorption on metal oxide surface (Zn), (b) Proton hopping in a nanocomposite structure made of water-bonded molecules.

III. Literature Based on Previous Works

Composite materials are assembled by merging and combining two or more materials with different and unique characteristics. The different materials in the composite work together to give the unique composite properties. Typically, in nanocomposite materials, one of the constituent materials usually exhibits a nanoscale structure in nanocomposite materials or nanoscale structures that repeat distances between the different phases that make up the material. Generally, the nanostructure composite formation improves the specific properties; mechanical strength, the conductivity of electricity, thermally and mechanically stability, resistance to chemical changes, and surface reactivity compared to the base material.

As far as sensing parameters such as sensitivity, reproducibility, response time and recovery time are concerned, the overwhelming majority of humidity sensors published previously perform excellently in one or two parameters. However, they continue to fall short in terms of overall performance across all parameters. Table 1 summarised the various composite structures, sensing materials and its effect to resistive type-based humidity sensor sensing performance in term of response/recovery time and sensitivity. The synergistic effect among each element of the heterojunction will compensate for the limitation of a single functional material and propose new sensing properties. Researchers have made necessary efforts to improve the heterojunction of ZnO and have demonstrated rapid electron transfer for material films.

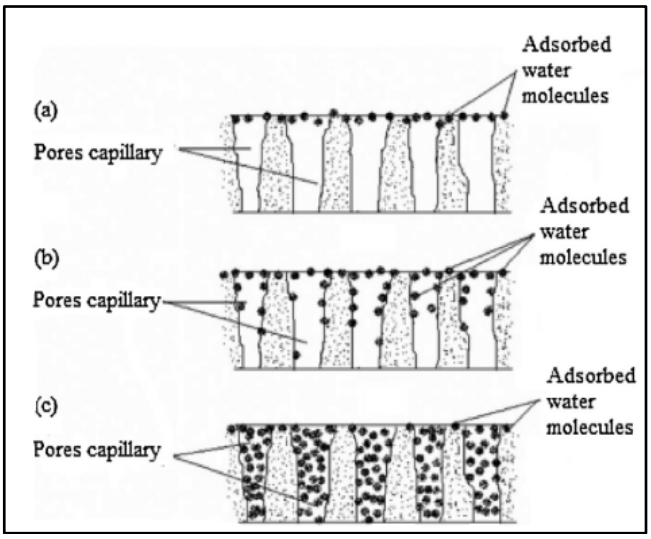

Fig. 4. (a) Surface adsorption, (b) Adsorption on capillary pore channel and, (c) Fully capillary condensation [33].

Chemical methods are used to generate nanostructured ZnO in the immense majority of situations, with physical approaches being used in a few cases. Chemical methods are relatively inexpensive and facile, but they may have problems with large-scale production, homogeneity, and repeatability. Physical techniques, in addition to chemical synthesis routes, have been effectively utilized for nanostructure production. The simplest physical method for producing zinc nanostructures is to oxidize zinc metal [55]. The majority of findings on ZnO nanostructure and composite production in this literature are based on a chemical synthesis technique in which zinc salts such as acetate, nitrate, or chloride are mixed with bases solution to initiate a reaction that results in the formation of zinc hydroxide (ZnOH). Consequently, this hydroxide (OH) is thermally oxidized to produce ZnO. The reaction is commonly accomplished in a solvent, and the final product is produced as powder either by washing and drying or by coating/absorbing on a substrate by thermal treatment.

The most frequent chemical synthesis technique used is precipitation, sol-gel, and hydrothermal. Controlled precipitation is indeed a broadly applied method of obtaining zinc oxide (powder and composite) because it allows for the production of a consistent product. Using a reducing agent, a rapid and spontaneous reduction of a zinc salt solution is achieved. The concentration of reagents, and the reaction temperature were controlled variables in this process [56]–[58]. By using precipitation method, well distributed particle size with a large surface area zinc oxide composite was generated [59]. On the other hand, the sol-gel approach for generating ZnO nanopowders composites has piqued interest due to its

simplicity, cost effectiveness, reliability, reproducibility, and very moderate synthesis conditions, which allow for surface modification of zinc oxide with specified organic chemicals [60], [61]. Hydrolysis and polymerization will turn the precursor solution into a sol. Heat treatment transforms the sol mostly into dense films. Meanwhile, the nanopowders were made from a colloidal sol that turned into a gel once the sol particles condensed [62]. The hydrothermal approach is easy and environmentally friendly because it does not involve the use of organic solvents or extra product processing such grinding or calcination. Crystal nuclei are generated as a result of heating and cooling, and they then grow into film or particles [63], [64]. The ability to carry out the synthesis at low temperatures, the variety of morphology of the resulting product were depending on the process parameter (temperature and pressure) and the composition of the initial mixture, the product high quality of crystallinity, and the high purity of the material obtained are just a few of the benefits of this process [55], [65]. Further modifications can be made by switching from thermal heating to microwave heating in an approach known as microwave assisted solvothermal synthesis. The primary advantage of these methods is that it uses microwaves to heat more efficiently than traditional heat transfer methods like as conduction and convection while maintaining higher homogeneity and reduced reaction times [66].

Ismail et al. reported that highly sensitive and extremely thin heterojunction films were prepared via a two-step solution-based conducted through a spin-coating and immersion methods for humidity-sensing application [41]. In their experimental setup, zinc acetate dihydrate are use as starting material for zinc oxide film deposition while tin (IV) chloride pentahydrate were use as tin layer starting material. Based on their findings, the average diameters of the ZnO and SnO_2 nanoparticles were 26 and 6 nm, respectively. According to their XRD analysis, the establishment of a heterointerface between SnO_2 and ZnO resulted in the ZnO crystallite size being reduced. With decreasing of the grain size and crystallite size, they suggested when deposited with SnO_2, the surface areas of the ZnO films were enhanced. Therefore, they conclude by reducing the deposition of SnO_2 to 3 min, the film resistance conclusively reduced to 0.40MΩ from 6.74 MΩ. Based on the humidity-sensing performance, the hetero-junction sensors were significantly dependent on deposition time, with the highest sensitivity (90.56) to humidity recorded for the sensors exposed to 3 min deposition time, which was drastically higher compared to bare ZnO. This is due to improvement of the electrostatic attraction of water molecules to the film surface by increasing the free carrier concentration in the film. They also suggested that the use of SnO_2/ZnO heterojunction has a great potential in humidity sensing applications.

Md Sin et al. were successfully fabricated humidity sensor utilizing sputtered ZnO-film-coated glass as substrate. They are using nanocubic based on ZnO and SnO_2 which prepared through sol–gel and immersion method [42]. Based on their experimental work, the cubic structure was synthesis through the ultrasonic sol–gel immersion using tin (IV) chloride pentahydrate and zinc chloride as starting material with specific controlled molar ratio of Zn:Sn and sodium hydroxide as mineralizer. By varying the molar of Zn, the nanocubic structured ZnO/SnO_2 were synthesis at difference molar ratio of Zn:Sn. They found that as the Zn molar increase the size of the cube is dramatically reduces. Corresponding to the ultra-violet (UV) emission and visible emission, respectively, the luminescence of nanocubic structured ZnO/SnO_2 was revealed around 400 and 600 nm. For humidity sensing performance, the best sensitivity of 104 times is logged for the composition of molar ratio 10:10 of nanocubic structured ZnO/SnO_2. With high response (411 s) and recovery times (98 s), apart from good repeatability and stability, they proposed that nanocubic structured ZnO/ SnO_2 have a great potential for humidity sensor application.

In other work by Ismail and his co-researchers, a novel two-step solution immersion method was employed to synthesis a distinctive heteronetwork composite which comprising of crystalline tin-doped zinc oxide nanorod arrays (SZO) and rutile tin oxide nanowires (SnO_2) for humidity sensing applications [43]. According to their experimental procedure, the coated glass substrate containing SZO was immersed into the solution containing tin precursor a low processing temperature and shorter immersion time. According to the surface morphology and cross-sectional images, they observed that the surface area and porosity channels of the film are increased by orienting SnO_2 nanowires in different directions on top of the SZO structures. They also mentioned that the average diameter and thickness of the SZO are roughly around 95 and 730 nm, respectively. Meanwhile, the SnO_2 nanowires diameter ranges from 60 nm to 120 nm, with nanowire length measured approximately 10 mm. They then proceed with the humidity sensing characteristic studies of the SZO/SnO_2-based humidity sensors at the humidity level ranging from 40% to 90% RH. They discovered, compared to SZO and pure SnO_2, the sensitivity of the SZO/SnO_2 composite based humidity sensor is far better (67.8) and exhibits high stability. Based on the FESEM images, the suggested that the significant expansion in surface area generated by the SnO_2 nanowire layer can be attributed for the outstanding SZO/SnO2 humidity sensing capability. Moreover, the pore channels seen between nanorod, and nanowire films provide additional water molecule adsorption sites. They also indicate with the formation of a heterojunction between two consecutive films helps to increase surface area, improve electron transfer, and reduce electron–hole recombination possibilities. Based on presented results, they concluded that the SZO/SnO2 heteronetwork unique architecture can improve the nanowires' humidity sensing competencies.

By utilizing gold nanoparticles (AuNPs) and black zinc oxide (ZnO) nanorods, Zhang et al. successfully prepared a highly sensitive self-powered humidity sensor has been realized through a sol-gel method [44]. The ZnO/AuNPs nanocomposite were produced by varying the amount of AuNPs added into precursors solution. Based on the structural characterization, the XRD reveals that both ZnO and ZnO/AuNPs display a wurtzite structure. Meanwhile, according to FESEM analysis, vertically alignment ZnO/AuNPs nanorods were successfully grown. Further analysis found that the nanorod possesses high uniformity and forms dense arrays with a smaller diameter compared to ZnO nanoparticles. Upon measurement, the diameter of pure ZnO is distributed from 60 nm to 200nm while for ZnO/AuNPs nanocomposite, the smaller diameter from 45 nm to 60 nm observed. This is due to the controlled growth of ZnO nanorods induced by the AuNPs. In term of band gap energy, all ZnO/AuNPs and pure black ZnO show lesser value than pure ZnO (3.34 eV). They also discover that amount of AuNPs is effectively influenced the band gap value of ZnO/AuNPs nanocomposites. For the humidity sensing experiment, using supersaturated aqueous solution that generated 11% to 95% RH at 25 °C, a controlled humidity environment was achieved. The humidity sensing measurements show that all ZnO/AuNPs humidity sensors have a significantly higher response compared to ZnO sensors. Meanwhile, compared to pure ZnO, the sensitivity of 6 mL AuNPs in ZnO/AuNPs nanorods display a change three orders higher with relative humidity (RH) ranging from 11% to 95% at room temperature. Their findings also highlighting that the best response and recovery time of the ZnO/AuNP nanocomposite is achieved with 6 mL AuNPs sample which measured at 5.6 s and 32.4 s, respectively. They concluded that the ZnO/AuNP nanocomposite with 6 mL AuNPs humidity sensor is reversible with fast response, good repeatability and stable. Finally, they suggested that the way to control the morphology of semiconductor nanomaterials is by construction of semiconductor/noble metal nanocomposite structured that eventually can be utilized to design a humidity sensor with high performance.

TABLE 1: SUMMARY OF COMPOSITED ZINC OXIDE BASED RESISTIVE TYPE HUMIDITY SENSOR

Sensing Material	Fabrication Method	Composite Structure	RH Range (%)	Response/ Recovery	Sensitivity	Comparison Parameter	Ref.
SnO_2/ZnO	Sol-gel and immersion	Bi-layer	40 – 90	-	90.56 [R_a/R_{rh}]	SnO_2 deposition time	[41]
ZnO/SnO_2	Sol-gel and immersion	Mixed nanostructure compound	40 – 90	411 s/98 s	109 [R_a/R_{rh}]	Zn:Sn molar ratio	[42]
SZO/ SnO_2	Sol-gel and immersion	Mixed nanostructure compound	40 – 90	-	67.8 [R_a/R_{rh}]	SZO	[43]
ZnO/Au	Sol-gel	Decorated nanostructures	11 – 95	5.6 s/32.4 s	-	AuNP ratio	[44]
ZnO/CuO	Microwave-assisted	Mixed nanostructure compound	10 – 98	158 s/426 s	1.4 [R_a/R_{rh}]	Annealing temperature	[45]
ZnO/ $ZnWO_4$	Precipitation	Mixed nanostructure compound	5 – 98	50 s/100 s	3416 [R_a/R_{rh}]	$ZnWO_4$:ZnO molar ratio	[46]
rGO/ZnO/Cu	Chemical bath deposition	Core-shell	11–97	19 s/42 s	-	NA	[47]
PVDF/ZnO	Hydrothermal	Decorated nanostructures	5 – 98	30 s/51 s	3417 [R_a/R_{rh}]	Individual PVDF and ZnO	[48]
ZnO/GaN	Spray deposition	Mixed nanostructure compound	12–96	7 s/13 s	161[R_a/R_{rh}]	ZnO	[49]
ZnO/ZrO_2	Chemical bath deposition	Mixed nanostructure compound	30–90	53 s/69 s	0.786 [R_a - R_{rh} /R_{rh}]	ZnO and different ZrO_2 amount	[50]

Using zinc acetate, copper acetate, 1-butyl-3-methyl imidazolium tetrafluoro borate and sodium hydroxide as the precursor materials, Ashok et al. were efficaciously synthesized ZnO/CuO nanocomposite using microwave-assisted technique using ionic liquid [45]. Setting the composition of ZnO and CuO to 1:1 wt.%, by using mortar and pestle, ZnO/CuO nanocomposite was prepared by manual grinding of a mixture containing ZnO and CuO. The structural morphology of the nanocomposite was studied by X-ray diffractometer (XRD), particle size analyzer (PSA) and scanning electron microscope (SEM), while Fourier transform infrared spectrometer (FTIR) and thermo gravimetric and differential thermal analyzer (TGDTA) were used to evaluate the chemical properties of the ZnO/CuO nanocomposite. The ZnO/CuO nano- composite was confirmed by XRD lies in average crystallite size in nano meter range with obtained

average particle size is between 28 nm and 34 nm for different temperatures. Highly porous nature of ZnO/CuO nanocomposite observed when the sample annealed at 600 °C. This is due when temperature increased the agglomeration takes place, depending on the agglomeration the particle size increased. The particle size increment causes the increasing of porosity. The Zn-O and Cu-O stretching bands were observed in FTIR. The strong peaks around 1600 cm^{-1} and 3460 cm^{-1} indicates that the stretching band of water molecules as well as hydroxyl groups which is on the surface of ZnO/CuO nanocomposite. Further analysis is carried out using TGDTA to analyze the weight losses of the sample and phase transformation. In both cases of annealing temperatures (500 °C and 600 °C) the weight losses were obtained as 1.7% and 1.4% for 500 °C and 600 °C, respectively. This weight decrement observed due to the increasing of annealing temperature. In term of humidity sensing performance, they recorded that the response time and recovery time of the sensing elements were logged as 172 s and 495 s for 500 °C annealing nanocomposite, whereas for 600 °C annealed sample, the response time and recovery time recorded as 158 s and 426 s, respectively. Therefore, the suggested that the humidity sensing property of the nanocomposite increased along with increasing of annealing temperature.

Composite consists of zinc tungstate ($ZnWO_4$) and zinc oxide (ZnO) nanoparticles were prepared via precipitation method for electrical and humidity sensing properties by Arularasu and Sundaram [46]. To achieve and fabricate $ZnWO_4$/ZnO composites, they mixed together the different moles ratios of $ZnWO_4$ and ZnO nanoparticles follow by sintering process in the form of the disc pallets. According to structural analysis findings, the pure phased of $ZnWO_4$ monoclinic wolframite tungstate is clearly identified in the diffraction peaks, while all of the ZnO peaks are in satisfactory correlation with wurtzite ZnO with the crystallite size was calculated to be 38 nm. As for morphological analysis, $ZnWO_4$/ZnO composites are distinguished by a porous structure and tiny grains with many inter grain pores. They indicated that the adsorption and condensation of water vapors are favored by this structure due to water vapour can penetrate the pores of porous metal oxide sensors, causing adsorption to occur in the pores. For humidity sensing performance characteristic, the controllable humidity surroundings were created by placing saturated salt solutions in a closed glass container at ambient temperature, which yielded 5% to 98% RH. They observed from these experimental values, with ratio of 40% $ZnWO_4$ to 60% of ZnO, the composite shows maximum sensitivity factor value of 3416 compared to other molar rations composite. That was because $ZnWO_4$/ZnO composites with 40:60 ratio possesses more pores and voids in their morphology than other composites which facilitating interaction with water molecules. The response time and recovery time of 40% $ZnWO_4$ to 60% of ZnO composite are 50 s and 100 s, respectively with a good long-term stability of DC resistance value overtime.

Kuntal et al. in their work reported that one-pot synthesis method was utilizing to prepare flower-shaped morphology nanocomposite that consists of metal (Cu) nanoparticles, a metal oxide (ZnO nanorods), and reduced graphene oxide (rGO) [47]. This nanocomposite is later used for the AC conductivity and humidity sensing response studies at room temperature. According to the morphology analysis, the nanocomposite shows confirmed that the ZnO nanorods consisted of pure wurtzite ZnO were present and these nanostructures resemble dahlia flowers. They mentioned the flower-shaped nanocomposite with overall diameters of about 1μm. This nanocomposite consisting of flower petal-like structures composed of rGO, ZnO, and Cu nanostructures. Kuntal and his teammates suggested that the conduction in the nanocomposite due to the hopping mechanism. This suggestion then further confirmed by studying the power law behavior of its AC conductivity. Based on experimental work at room temperature, due to adsorption of water molecules on the surface of nanocomposite, they observed the nanocomposite showed an increasing in sensing response and a decreasing in resistance with an increase in relative humidity (from 11 to 97%). The calculated sensing response revealed that the nanocomposite shows a maximum sensing response of 97.79% in the broad humidity range of 11–97% RH. Apart from that, the response and recovery times of the nanocomposite were 19 s and 42 s, respectively when tested from 11 to 97% RH and moved from 97% RH back to 11% RH. Apart from that, they conclude that the humidity sensor based rGO, ZnO, and Cu nanocomposite has a low humidity hysteresis, good repeatability and a consistent humidity sensing ability, making it an excellent choice for humidity-sensing applications.

PVDF/ZnO nanocomposite was prepared as a humidity sensor by Arularasu et al. using a simple hydrothermal method [48]. In their work, PVDF, an aliphatic polymer with substantial hydrophobic groups, has been formulated as a hydrophilic humidity-sensitive material with the incorporation of ZnO nanoparticles as resistant transduction layers. Based on morphology analysis, the observed that PVDF/ZnO nanocomposite exist in 2D replicated structure with rod-like nanocomposite, rough surface and uniform morphological. With addition of PVDF in ZnO nanoparticles, it also helps to achieve a rough morphology with grain sizes ranging from 25 to 75 nm. The humidity sensing properties is observed having improvement due to the porous structure and uniformly distributed structure that provide more water molecules adsorption and desorption sites across the humidity sensor surface. To study and observed the humidity sensing performance and properties, they were using saturated salt solutions to achieve the relative humidity range from 5% to 95% at 35°C. According to experimental results, its shows and confirmed that the PVDF/ZnO nanocomposite humidity sensor has a 3 order of magnitude greater variation than pure PVDF and has better linearity than pure ZnO nanoparticles. The calculated sensitivity factor for PVDF/ZnO nanocomposite humidity sensor were 3417. As comparison, the sensitivity

factor for pure PVDF and ZnO nanoparticle were 188 and 363, respectively. The increased humidity sensitivity was most likely due to the high surface region, which offers more water adsorption sites in the hybrid nanocomposite, according to the experimental results. Surface area investigation revealed that ZnO nanoparticles had a lesser surface area than ZnO and PVDF/ZnO nanocomposite. As a result, the increased surface area of the PVDF/ZnO nanocomposite is anticipated to improve humidity sensing activity. Apart from that, morphology such rod like structure of the PVDF/ZnO nanocomposite, decreases in the particles size and have more adsorption site completely help to provide fast response and recovery time logged at 30 s and 51 s, respectively. As a result, the nanocomposite's swift response and recovery at various RH levels, as well as its strong stability at room temperature, make it ideal for use as a humidity sensor. Finally, based on obtained results, they conclude that these findings pave the way for a new approach to high-performance and promising 2D nanocomposite materials in the production of sensitive resistive type humidity sensors.

A resistive-type humidity sensor based on a ZnO/porous GaN (ZnO/PGAN) heterojunction as sensing material were fabricated by Wang and his co-workers [49]. In their works, using hydrothermal method, the ZnO disks were synthesized by mixing the equimolar amounts of sodium dodecyl sulfate and cetyltrimethylammonium bromide into specific amount zinc nitrate hexahydrate with addition of sodium hydroxide. Then by managing the spraying speed to balance the spraying and solvent evaporation time, a thick micro-structured zinc oxide (ZnO disks) film was gradually deposited on the PGAN substrate. By using X-ray diffraction and scanning electron microscopy, the structure and surface morphology of the ZnO/PGAN were characterized. According to the SEM image, the surface of the ZnO/PGAN device is packed with well dispersed uniform hexagonal structures of ZnO disks. Based on measurement done, the diameter of the ZnO disks is in range from 150 to 220 nm. Based on investigation thru photoluminescence spectra and I–V characteristics curve, they observed that the heterojunction demonstrated an exceptional diode nature. In term of humidity sensing performance analysis, the RH-controlled environment was obtained by using saturated salt solutions for range of 12% to 96% RH and shows high sensitivity (161.0) for a broad mentioned humidity range. They claimed that the excellent transport capability of ZnO/PGAN lead to increased electron transfer, resulting in high sensitivity. In the range of 12–96% RH, a fast-sensing response time as low as 7 s and a recovery time of 13 s were achieved. The stability of the ZnO/ PGAN heterojunction humidity sensor under various humidity level and they observed for continuous sensing of more than 2 hours, the device maintains its stable performance and sensitivity with very minimal drift and hysteresis, indicating its stability and durability. Apart from that, after four cycle tests, the sensor demonstrates extremely good repeatability with 96% baseline remains maintains unaffected. Finally, they conclude and stated the versatility of a ZnO/PGAN-based humidity sensor is highlighted by its simplistic nature.

In their present investigation, Arote and his co-worker, were successfully synthesized zinc oxide-zirconium oxide (ZnO-ZrO_2) nanocomposite-based structure by ultrasonic assisted wet chemical procedure [50]. The composite nanoparticle structural, optical and humidity sensing properties were investigated. According to XRD and EDS analysis of the sample with addition of certain amount of ZrO_2, the presence of ZrO_2 diffraction peaks in the XRD spectra confirm the formation of ZnO-ZrO_2 composite. Moreover, the phase formation and phase purity of the prepared composite were aligned with the respective standard data. Based on Williamson-Hall method, they observed with ZrO_2 content is increased in ZnO-ZrO_2 composites, the crystallite size gradually decreased. With this finding, they suggested this correlation will cause the surface area to volume ratio of the composite structure will increased and may be attributed to small grain growth during synthesis procedure. This argument is further supported by the SEM micrographs image. According to SEM evidence, with increasing concentrations of ZrO_2, the morphology of the primary compound ZnO changes dramatically. Micropores can even be seen between the grains, and their sizes show a definite relationship with ZrO_2 concentration. The diameters of micropores were found to decrease as ZrO_2 concentration increased, which is relevant since micro-pores play an important role in sensing properties of nanomaterials. The humidity sensing performance of all samples was validated, and the impact of ZrO2 concentration in ZnO-ZrO_2 composites on sensitivity, response, and recovery time was thoroughly investigated. The sensitivity of ZnO-ZrO_2 composites increased from 56.05 percent to 78.26 percent as the amount of ZrO_2 was increased. However, when the amount of ZrO_2 in the composite material increases, the sensitivity of the material decreases. They were able to prove that the surface morphology of nanomaterials affects the humidity sensing properties significantly. The ability of water molecules to condense on the surface of nanomaterials is governed by the material's porosity, which is further influenced by the presence and density of micropores. According to humidity sensitivity performance calculation, small micropores have an advantage over vast surface areas in terms of chemisorbed layer development. Due to presence of the micropores, the fast response time of 53 s and recovery time of 69 s were recorded for the sample with addition of 0.4 g ZrO_2, which can be attributed to the water molecules at its surface adsorbing and desorbing at a quicker rate.

According to extensive research presented above, surface decorating of ZnO films employing the compositing approach is critical, particularly for humidity-sensing applications. Incorporating composite layers to nanostructured ZnO has been shown to be capable of altering the properties of pristine ZnO while also improving the performance of numerous devices, most notably humidity sensors. Metal oxides elements (SnO_2, AuNP, CuO, $ZnWO_4$, and ZrO_2) are recognised to have

several advantages, including a high energy gap, outstanding physical and chemical properties, and a highly sensitive material make them suitable for this purpose. When integrated with ZnO, these metal oxide nanostructures increased surface area, contribute to high-quality surface structure and porosity, provided substantial concentrations of oxygen vacancies, improved electron transport, and reduced the potential of electron-hole recombination. As mentioned in Table 1, composited this material with ZnO through composite approach improved humidity sensing performance of the sensor in one or more area. In case of graphene or reduced graphene oxide (RGO), once composited with ZnO, the hydrophilicity of the material improves dramatically [18]. This is attributed to the highly conductive sp^2 carbon atom sheet, which can operate as an anchor to facilitate electron transport with other sensing materials [51]. Additionally, RGO films not only exhibit high surface-to-volume ratios, but also acquire an elevated concentration of surface vacancies and hydrophilic carboxylic functional groups, enabling RGO to easily absorb water molecules from their environment [52], [53]. Aside from that, RGO has proven to be an incredibly exciting electro-active element due to its distinctive characteristics which include high charge carrier manoeuvrability, outstanding conductivity, high mechanical strength, and extremely high thermal conductivity due to the ability to retain and release energy via charge separation at the electrode and electrolyte interface. [54].

IV. Conclusion

In this presented review, we concentrated on latest developments of ZnO composite structure for resistive type humidity sensor applications. Additionally, the detailed working principle of the positive conduction mechanism for resistive type humidity sensor also have been discussed. Throughout this review, we have shown that a vast variety of diverse morphologies and structures can be synthetized, each with unique sensing properties for a broad spectrum of relative humidity level (%RH). The fabrication of structures with various shapes, morphologies, and dimensions was made possible by integrating ZnO with other metals, metal oxide, graphene-based material, and polymer. This assessment of the literature reveals that a substantial percentage of the research community has been working on the synthesis of composite structure using uncomplicated and straightforward techniques such as hydrothermal, sol-gel, bath deposition, and solution immersion. The mentioned techniques as synthesis procedures is benefited from the simplicity of the technique and the availability of various precursor materials. According to previous studies, ZnO has been composited of numerous types of elements such as metal, metal oxide, carbon-based material, and polymer that is created using various synthesis methods. Applying composite layers (or other types of composite structures) to ZnO structures has been shown to be effective in improving the properties of pristine ZnO and thus significantly improving the performance of resistive type humidity sensors.

Finally, although the composite nanostructures metal oxide device has the potential to revolutionize humidity sensing, there are still numerous discoveries and experiments that need to be conducted in order to fully comprehend the capabilities of these materials. As a result, the underlying final purpose of this review was not only to describe the state of the art, but also to stimulate and inspire readers' curiosity, causing them to investigate this specific category of nanomaterials in greater depth.

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