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Recent Advances of SnO_2-Based Sensors for Detecting Fault Characteristic Gases Extracted From Power Transformer Oil

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Tin oxide SnO_2-based gas sensors have been widely used for detecting typical fault characteristic gases extracted from power transformer oil, namely, H_2, CO, CO_2, CH_4, C_2H_2, C_2H_4, and C_2H_6, due to the remarkable advantages of high sensitivity, fast response, long-term stability, and so on. Herein, we present an overview of the recent significant improvement in fabrication and application of high performance SnO_2-based sensors for detecting these fault characteristic gases. Promising materials for the sensitive and selective detection of each kind of fault characteristic gas have been identified. Meanwhile, the corresponding sensing mechanisms of SnO_2-based gas sensors of these fault characteristic gases are comprehensively discussed. In the final section of this review, the major challenges and promising developments in this domain are also given.

Keywords: tin oxide, gas sensors, fault characteristic gases, power transformer oil, sensing properties, sensing mechanism

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INTRODUCTION

Power transformers are one of the most important apparatuses in power systems, and their reliability is extremely vital to ensure the stable system operation (Zheng et al., 2018). Thermal and electrical faults in oil-filled power transformers may produce typical fault characteristic gases including hydrogen H_2 (David et al., 2018), carbon monoxide CO (Joseph, 1980; Uddin et al., 2016; Zhou et al., 2018b), carbon dioxide CO_2 (2015; Dan et al., 2016; Iwata et al., 2017; Zhang et al., 2017), methane CH_4 (Sedghi et al., 2010), acetylene C_2H_2 (Qi et al., 2008), ethylene C_2H_4, and ethane C_2H_6. These typical fault characteristic gases could be dissolved in transformer oil or accumulate as free gases if produced rapidly in large quantities. Therefore, detection and analysis of the species, quantities and generation rates of these fault gases presented in the fluid allow for the identification of power transformer fault types such as corona, sparking, overheating, and arcing.

During the past few decades, dissolved gas analysis has been developed to detect the latent faults of oil-immersed power transformers (Morais and Rolim, 2006; Zhan et al., 2015; Uddin et al., 2016; Gong et al., 2018). Gas sensing detection technology is the core of dissolved gas analysis. Different types of sensing technologies have been reported in previous studies for detecting typical fault characteristic gases extracted from transformer oil, such as metal oxide semiconductor gas sensors (Zhu and Zeng, 2017a; Chen et al., 2018), catalytic combustion sensors (Liu et al., 2011),

fuel cell sensors (Modjtahedi et al., 2016; Tonezzer et al., 2017), and optical sensors (Trieu-Vuong et al., 2016; Paliwal et al., 2017). Given the remarkable advantages of simple fabrication process, low maintenance cost, fast response and recovery, long service life, and so on, metal oxide semiconductor materials like SnO_2 (Choi et al., 2014; Zeng et al., 2014), ZnO (Zhou et al., 2017; Zhu and Zeng, 2017b; Zhu et al., 2017), TiO_2 (Zeng and Liu, 2010; Zhang et al., 2018b) and In_2O_3 (Cao et al., 2015) have received scientific and technological importance for many years and are widely used to detect these gases. Among them, as the most widely used gas-sensitive material, SnO_2 is currently the main sensing materials used in experimental research and commercial application for detecting typical fault characteristic gases extracted from power transformer oil (Zhang et al., 2010; Zhou et al., 2018a).

Herein, the first section of this review will briefly outline the preparation of the currently used SnO_2 sensing materials, the microstructure morphology, and doping modification of SnO_2 sensing materials for detecting typical fault characteristic gases extracted from power transformer oil. The second section addresses the main gas sensing mechanisms of SnO_2 gas sensors for these gases. The third section presents the conclusions, focusing on future challenges and potentialities associated with SnO_2-based gas sensors for detecting these typical fault characteristic gases.

SENSING PERFORMANCES OF SNO_2-BASED SENSORS TO FAULT CHARACTERISTIC GASES EXTRACTED FROM POWER TRANSFORMER OIL

For detecting and analyzing typical fault characteristic gases extracted from power transformer oil with SnO_2-based gas sensors, the most important concerns are the sensor sensitivity, selectivity, and repeatability (Fan et al., 2017). Combined with the research concerns mentioned above, this section briefly summarizes recent progress in the application of SnO_2 sensing materials to detect typical fault characteristic gases dissolved in transformer oil. The gas sensing performance of some modified SnO_2 gas sensors are listed in **Table 1**.

T_{oper}, temperature at which the best sensor performance, usually in terms of the highest response toward target gas, could be obtained.

S, gas sensing response of a SnO_2-based sensor to target gas, which is defined as S =Ra/Rg for reducing gas and S = Rg/Ra for oxidizing gas (where Ra and Rg are the resistance of the sensor in air and in the test gas, respectively).

It is known that the metal oxide semiconductor SnO_2 is a promising material for sensing the typical fault characteristic gases extracted from power transformer oil. Nevertheless, the sensing performance of the proposed sensors considerably depends on the preparation method and surface structure of the sensing materials and the dopants. The structure of modern SnO_2 gas sensors could be classified into two main types including thick/thin films and nanoparticles. The preparation methods of the sensing materials mainly include the hydrothermal method, the sol-gel method, the electrospinning technique, chemical vapor deposition and so on. Different preparation methods could affect the morphology of the sensing materials and further change their gas sensing properties (Long et al., 2018; Zhang et al., 2018a; Zhou et al., 2018c). Noble metals or metal oxide doped on SnO_2-based sensors can play an important role in accelerating the sensing process and improving the gas sensor performances.

In power transformers, H_2 is generated from the thermal decomposition of oil at high temperatures, which is a serious problem for transformer oil (Uddin et al., 2016). Various high-efficacy and workable SnO_2-based sensors have been recently introduced for detecting H_2. It can be seen from the data in **Table 1** that various metal particles such as Co (Liu et al., 2010), Au (Yin and Tao, 2017), and Pd (Nguyen et al., 2017) have been added to SnO_2-based sensors to enhance H_2 gas sensing performance. Among them, the Au-loaded SnO_2 sensor can detect H_2 down to 1 ppm, which is a good property for detecting low concentrations of H_2 dissolved in transformer oil (Yin and Tao, 2017). K. Inyawilert reported that a SnO_2 sensing film with an optimal Rh-doping level of 0.2 wt% exhibited an ultra-high response of 22,170 and a short response time of 6 s toward 30,000 ppm H_2 at an optimum operating temperature of 300°C. In addition, the proposed Rh-doped SnO_2 sensor displayed good H_2 selectivity against NO_2, SO_2, C_2H_4, C_3H_6O, CH_4, H_2S, and CO (Inyawilert et al., 2017). Except for nanoparticles embedded in the metal oxide matrix, the morphology of the SnO_2 materials could be applied to improve the H_2 sensing properties, and low dimensional nanostructures have attracted increasing attention. Nguyen Kien et al. introduced the on-chip growth of SnO_2 nanowire-based gas sensors to detect low concentrations of H_2 gas, which responded with 2.6 for 20 ppm H_2 gas at 400°C (Nguyen et al., 2017). Taken together, these findings suggest that the Au-loaded SnO_2 gas sensor is the most promising candidate for detecting low concentration H_2 extracted from power transformer oil.

The insulating paper, pressboard and wood blocks in power transformers contain a large number of anhydroglucose rings as well as weak C-O molecular bonds and glycosidic bonds that are thermally less stable than the hydrocarbon bonds in oil. These bonds could decompose to CO and CO_2 at lower temperatures when potential faults occur. As shown in **Table 1**, Pt-doped SnO_2 thick film (Chen et al., 2018), ZnO-SnO_2 nanoparticles (Chen et al., 2015), and Au-doped SnO_2 yolk-shell nanospheres (Wang et al., 2013) could be potential candidates for CO sensors. Among them, Wang et al. compared the gas sensing properties of Au@SnO_2 yolk-shell nanospheres with that of hollow SnO_2 nanospheres. The sensor fabricated with the Au@SnO_2 yolk-shell nanospheres showed lower operating temperature (210°C), lower detection limit (5 ppm), faster response (0.3 s), and better selectivity to CO gas (Wang et al., 2013), which could be attributed to the catalytic effect of Au and enhanced electron depletion at the surface of the Au@SnO_2 yolk–shell nanospheres. Moreover, SnO_2 loaded with Pd nanoparticles may be another kind of promising material for CO gas sensing (Zhou et al., 2018d). Yuasa et al. prepared PdO-loaded SnO_2 nanoparticles by the reverse micelle method and reported that the 0.1 mol% PdO-loaded SnO_2 sensor exhibited a high gas response value of 320

TABLE 1 | Comparison of the representative SnO_2 based sensors for fault characteristic gases extracted from power transformer oil.

Target gas	Sensing material	Synthesis method	Morphology	Detection range (ppm)	$T_{oper.}$ (°C)	S/ppm	Selectivity	References
H_2	1 wt% Co-doped SnO_2	Electrospinning	Nanofibers	100–25,000	330	24/100	H_2 > CO	Liu et al., 2010
	1 mol% Au-doped SnO_2	Sol-gel	Nanoparticles	1–5,000	400	48/5,000	H_2 >CO	Yin and Tao, 2017
	Pd-decorated SnO_2	Chemical vapor deposition	Nanowires	10–100	350	7.1/100	H_2 >CO_2	Nguyen et al., 2017
CO	1.5% Pd-doped SnO_2	Co-precipitation	Thick-film	100–1,000	260	6.59/400	–	Chen et al., 2018
	ZnO–SnO_2 nanoparticles	Typical hydrothermal	Nanoparticles	40–160	300	13/100	–	Chen et al., 2015
	Au@SnO_2	Hydrothermal deposition	Yolk–shell nanospheres	5–100	210	30/50	CO>H_2 >C_2H_4 > CO_2	Wang et al., 2013
CO_2	$LaFeO_3$/SnO_2	Sol-gel	Thick film	–	250	2.72/4,000	–	Zhang et al., 2017
	LaOCl-doped SnO_2	One-step electrospinning	Nanofibers	100–20,000	300	3.7/1,000	–	Xiong et al., 2017
CH_4	20 mol% Pt-SnO_2	Electrospinning	Nanofibers	1–1,000	350	1.11/1	–	Lu et al., 2018
	Pt-SnO_2	Wet chemical	Thin film	1000–10,000	400	1.55/1,000	–	Min and Choi, 2005
C_2H_2	Sm_2O_3-doped SnO_2	Sol–gel	Nanoparticles	1–5,000	180	63.8/1,000	C_2H_2 >CO>CH_4 >H_2	Qi et al., 2008
	rGO-Loaded SnO_2	Hydrothermal	Nanoparticles	0.5–500	180	12.4/50	C_2H_2 >CH_4 >H_2 > CO>CO_2	Jin et al., 2017
C_2H_4	silicalite-1 layer coated on SnO_2	Ultrasonic spray pyrolysis technique	Thin film	2–70	350	2.21/8	–	Jadsadapattarakul et al., 2010
	SnO_2	R.F. magnetron sputtering	Thin film	10–100	300	10.43/100	–	Ahn et al., 2010
C_2H_6	5 wt% Pd-doped SnO_2	Hydrothermal	Nanoparticles	5–100	400	5.89/100	–	Chen et al., 2013a

to 200 ppm CO gas (Yuasa et al., 2009). Chen et al. synthesized Pd-doped SnO_2 nanoparticles using a co-precipitation method, and the 1.5 wt% PdO decorated SnO_2 presented the largest gas-sensitive response of 6.59 at 260°C in 400 ppm CO atmosphere (Chen et al., 2018). Another study reported that Pd-modified nanocrystalline SnO_2 displayed a fairly high and reversible CO response (2–100 ppm) at room temperature (Marikutsa et al., 2010). Yin et al. prepared Pd-loaded and Fe-doped SnO_2 by the sol-gel method. The composite with 10 mol% Fe and 0.2 mol% Pd had the highest sensitivity and selectivity to CO in the range of 200–3,000 ppm at 350°C, and the response value to 2,000 ppm CO was increased 13 times compared with pure SnO_2 (Yin and Guo, 2014). Therefore, Pd is a highly effective catalyst for improving the sensing performance of SnO_2-based sensors to CO gas (Xiong et al., 2017).

Due to their high chemical stability, conventional binary metal oxides have very low sensitivity to chemically inert gases such as CO_2 (Korotcenkov and Cho, 2017). However, it was reported that La-doped SnO_2 nanocomposites can be used for CO_2 sensing (Kim et al., 2000). And the 8% LaOCl-SnO_2 nanofibers exhibited an optimal response of 3.7 toward 1,000 ppm CO_2 at 300°C with response/recovery times of 24/92 s (Xiong et al., 2017). Karthik et al. reported the sensing properties of tin oxide (SnO_2) and zinc oxide (ZnO) thin films deposited onto macroporous silicon (PS) substrates to CO_2 and found that the obtained SnO_2/PS films showed the highest sensing response of 19 to 15 ppm CO_2 gas (Karthik et al., 2018).

For hydrocarbons, namely, CH_4, C_2H_2, C_2H_4, and C_2H_6, when high energy discharge occurs, such as arcing, in transformer oil, the content of C_2H_2 is relatively high. At low temperature thermal faults ($T < 300$°C), the contents of CH_4 and C_2H_6 tend to be high. C_2H_4 is the main content of hydrocarbon while at high temperature thermal faults ($T > 300$°C) (Fan et al., 2017). For detecting these hydrocarbons, an online monitoring system based on a SnO_2-based gas chromatographic detector for assessing the running condition of a power transformer was developed (Fan et al., 2017). Qi et al. fabricated a C_2H_2 sensor based on 6 wt% Sm_2O_3-doped SnO_2, whose gas response to 1,000 ppm C_2H_2 could reach 63.8, 16.8 times larger than that of pure SnO_2 (Qi et al., 2008).

Moreover, Jin et al. reported that reduced graphene oxide (rGO)-loaded SnO_2 hybrid nanocomposite showed high sensor response (12.4 toward 50 ppm), fast response-recovery time (54 and 23 s), low detection limit (1.3 ppm), good linearity, excellent selectivity and long-term stability to C_2H_2 (Jin et al., 2017). These results indicated that rGO would be an effective addition to enhance the sensing properties of SnO_2-based sensors to C_2H_2 and make a contribution to developing a ppm-level gas sensor for on-line monitoring of C_2H_2 gas extracted from transformer oil.

Sensors for CH_4 detection have also been widely studied and are partly summarized in **Table 1**. The 20 mol% Pt-SnO_2 nanofibers exhibited excellent CH_4 sensing properties over a temperature range of 100–350°C, and an obvious response of 1.11 to 1 ppm CH_4 was measured at 350°C (Lu et al., 2018). Koeck et al. found a device prepared by ultra-long single crystalline SnO_2-nanowires, which was able to detect a few ppm of CO and CH_4 at the operating temperature of 200–250°C (Koeck et al., 2009). Chen et al. successfully synthesized Co-doped SnO_2 nanofibers via an electrospinning method and reported that the Co-doped SnO_2 nanofiber sensor exhibited a high response of 30.28 toward 50 ppm CH_4 at 300°C (Chen et al., 2013b). For C_2H_4 and C_2H_6 detection, only a few sensors including SnO_2 thin films and Pd-doped SnO_2 nanoparticles are found to be effective. Jadsadapattarakul et al. reported that the sensing response, response and recovery time of SnO_2 thin film sensors for selective detecting of C_2H_4 gas could be improved by coating a layer of [010] highly preferred-orientation silicalite-1 polycrystals (Jadsadapattarakul et al., 2010). Ahn et al. obtained SnO_2 thin films by R.F. magnetron sputtering to fabricate high performance C_2H_4 gas sensors (Ahn et al., 2010). Chen et al. discovered that the 5 wt% Pd-doped SnO_2 sensor could detect C_2H_6 at 400°C, and the sensor exhibited the largest gas-sensitive response of 5.89 toward 100 ppm C_2H_6 (Chen et al., 2013a).

SENSING MECHANISMS OF SNO_2-BASED SENSORS TO FAULT CHARACTERISTIC GASES EXTRACTED FROM POWER TRANSFORMER OIL

It is well agreed that the sensing mechanism of SnO_2 gas sensors is the change in conductivity of the metal oxide layer caused by the interaction with the surrounding atmosphere (Ducéré et al., 2012; Zeng et al., 2013; Korotcenkov and Cho, 2017). When exposed to air, oxygen molecules would be adsorbed on the surface of the SnO_2 nanostructures and capture electrons from the conduction band of SnO_2 to generate chemisorbed oxygen species (O^-, O^{2-}, and O_2^-, depending on temperatures) (Shahabuddin et al., 2017). At low temperatures (below 150°C), oxygen molecules exist in the form of molecular ions O_2^-, which would change to atomic ions O^- (150–400°C) and O^{2-} (more than 400°C) as temperatures rising (Punginsang et al., 2017). The chemical adsorption process can be explained by the following reactions:

$$O_{2(\text{gas})} \Leftrightarrow O_{2(\text{abs})} \tag{1}$$

$$O_{2(\text{abs})}+e^- \Leftrightarrow {O_2}^-{}_{(\text{abs})} \tag{2}$$

$${O_2}^-{}_{(\text{abs})} + e^- \Leftrightarrow 2{O}^-{}_{(\text{abs})} \tag{3}$$

$${O}^-{}_{(\text{abs})} + e^- \Leftrightarrow {O}^{2-}{}_{(\text{abs})} \tag{4}$$

As the electrons transfer from the conduction band of SnO_2 to the chemisorbed oxygen, the electron concentration and electrical conductivity of the SnO_2 film decrease. When the SnO_2 film is exposed to typical fault characteristic gases, the reducing gas would react with the chemisorbed oxygen species, thereby releasing electrons back to the conduction band with increasing electrical conductivity. The sensing mechanisms of the SnO_2 sensor sensing these fault gases can be explained from the following reaction paths, where O^- is taken as an example (Samerjai et al., 2012).

$$H_2+{O}^-{}_{(\text{ads})} \rightarrow H_2O+e^- \tag{5}$$

$$CO+{O}^-{}_{(\text{ads})} \rightarrow CO_2+e^- \tag{6}$$

$$C_2H_2+{O}^-{}_{(\text{ads})} \rightarrow 2C+H_2O+e^-+\text{heat} \tag{7}$$

$$CH_4+2{O}^-{}_{(\text{ads})} \rightarrow 2H_2O + CO_2+2e^- \tag{8}$$

$$C_2H_4+2{O}^-{}_{(\text{ads})} \rightarrow 2H_2O + 2C + 2e^- \tag{9}$$

$$C_2H_6+3{O}^-{}_{(\text{ads})} \rightarrow 3H_2O + 2C + 3e^- \tag{10}$$

Dopants added to SnO_2-based gas sensors can accelerate the reaction process mentioned above and improve the sensing performance of gas sensors. The doped catalysts can enhance the sensing performance of a sensor in two ways, namely, chemical sensitization and electronic sensitization (Lin et al., 2017). The high surface area structures such as porous structures and hollow structures could accelerate gas diffusion on the material surface and then shorten the response time.

On the other hand, to understand the sensing mechanism of SnO_2-based materials at the atomic and quantum levels, based on the framework of Density Function Theory (DFT), Li et al. established a SnO_2 surface model, gas molecular models and adsorption models. A first principles calculation using the Cambridge Sequential Total Energy Package (CASTEP) program was performed, and the total energies, electronic structures and adsorption properties were investigated in detail. Theoretical calculations provided a qualitative explanation of the sensing properties of the fabricated SnO_2-based sensor to various fault characteristic gases (CH_4, C_2H_6, C_2H_4, and C_2H_2) extracted from power transformers (Li et al., 2017). Zeng et al. performed a first principles calculation to investigate how H_2 gas interacts with the SnO_2 (110) surface and the effect of metallic ions on the gas response of SnO_2. Based on the theoretical calculation, it was reported that the Pd-doped SnO_2 (110) surface could adsorb more H_2 gas and receive larger electrons from adsorbed H_2 molecules (Zeng et al., 2011). Other research involving a

possible CO sensing mechanism for Pd-doped SnO_2 sensors was investigated with first principles calculations (Chen et al., 2018), and the theoretical results demonstrated that CO molecules can grab O from the pre-adsorbed oxygen on the Pd^4 cluster or the PdO cluster on the SnO_2 (110) surface. These processes may play an important role in CO sensing for Pd-doped SnO_2 (Chen et al., 2018). In particular, theoretical calculations indicated that CO_2 molecules cannot be adsorbed onto the stoichiometric SnO_2 (110) surface or SnO_2 (110) surface pre-adsorbed by O_2^- and O^- in dry air. However in wet air, CO_2 could react with O of pre-adsorbed OH^-, bringing about the formation of carbonates containing $(CO_3)^{2-}$ and the dissociation/movement of surface OH^- groups, accompanying the release of electrons from CO_2 to the SnO_2 surface (Wang et al., 2016).

CONCLUSION AND PERSPECTIVE

In this mini-review, SnO_2-based gas sensors for detecting typical fault characteristic gases extracted from power transformer oil have been briefly summarized. The analysis shows that the detection performances of SnO_2-based gas sensors can be obviously enhanced with dopant addition or increasing the surface area of the sensing materials. Despite achieving good results, there is still much room for further development. First of all, due to the cross-sensitivity between these fault characteristic gases, selectivity is a large challenge for developing high performance SnO_2-based sensors to detect these gases independently. In the future, to promote engineering applications, a combination of SnO_2-based gas sensors and gas chromatography should be further studied for multi-component detection of these gases. For further development of the gas-sensing performances, the high surface area structures such as hollow and hierarchical nanostructures could be prepared to obtain more active sites for gas diffusion, and different types of dopant elements should be examined. Furthermore, the gas sensing mechanisms are still imperfect and controversial, which cannot effectively guide development of novel SnO_2-based sensors and limit the prevailing application of these sensors to fault characteristic gases extracted from power transformer oil. Further research should focus on determining a satisfying mechanism model of SnO_2-based sensors to provide a guide for future work.

AUTHOR CONTRIBUTIONS

All authors listed have made substantial, direct and intellectual contributions to the work and approved it for publication.

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