

## Fe-modification on the surface of SnO<sub>2</sub> nanoparticles for high performance gas sensor

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(酸化スズナノ粒子表面の鉄イオン修飾によるガスセンサの高性能化に関する研究)

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## 論 文 内 容 の 要 旨

The development of gas sensors has been receiving a great deal of attention as an essential device in various fields. Especially, semiconductor gas sensors are enabled easy access to the wide applications by attractive merits such as low fabrication cost, small unit size and low power consumption. But the presence of moisture in measurement atmosphere gives uncertainties for the gas detection using semiconductor gas sensors. In order to solve this problem, many studies have been reported on doping and loading methods to enhance gas sensing performance in humid atmosphere. However it have been not clarified the reason why these methods are useful for gas sensing from viewpoint of the gas sensing mechanism. Therefore it is required new design of gas sensing materials to minimize the influence of moisture.

In this study, the surface modification has been successfully designed to enhance gas sensing performance and it was carried out on SnO<sub>2</sub> nanoparticles by one-step method using HF/Fe nitrate solution. Neat and Fe-modified SnO<sub>2</sub> sensor devices were investigated in terms of oxygen adsorption properties, humidity effect and gas sensing performance to H<sub>2</sub> at various working temperatures. Fe-modified SnO<sub>2</sub> sensor gave O<sup>2-</sup> adsorption species, low humidity dependence and high response to H<sub>2</sub> according to electrical measurement results in dry and humid atmospheres. These behaviors were considered from the perspective of the role of Fe ions on the surface and donor density. Furthermore gas sensing properties were investigated using H<sub>2</sub>O/D<sub>2</sub>O exchange, monitored by DRIFTS, and performed in pure air and in the presence of H<sub>2</sub> in air. Absorbance spectra and sensor responses of neat and Fe-modified SnO<sub>2</sub> were compared to understand better the role of Fe-modification in reducing humidity induced influences. High H<sub>2</sub> response of Fe-modified SnO<sub>2</sub> sensor in humid atmosphere was discussed in the light of electric aspects of water vapor and oxygen ionosorption on the surface.

Chapter 1 is summarized the background of this study and the state of research in related fields.

Chapter 2 is described about electrical properties in dry atmosphere in terms of oxygen adsorption behaviors and gas response to H<sub>2</sub> in order to confirm oxygen adsorption species and gas sensing performance. The surface modification of SnO<sub>2</sub> nanoparticles with Fe ions was involved in the result of O<sup>2-</sup> species as a oxygen adsorption, judging from the linear relationship between electric resistance and oxygen partial pressure. It was also caused high response and fast reaction speed to H<sub>2</sub>. It seems that Fe ions are strongly concerned in decreasing donor density and volume depletion state by surface

electron trapping. As a result, surface modification by Fe/HF treatment was demonstrated potential to enhance gas response and stability in dry atmosphere.

Chapter 3 is ranged over electrical properties in humid atmosphere to verify effect of humidity and change of oxygen adsorption species. These factors were explained with regard to oxygen adsorption, humidity dependence and gas response to H<sub>2</sub>. The Fe-modified SnO<sub>2</sub> sensor devices were shown the characteristics of low humidity dependence from the results of oxygen adsorption and humidity dependence in comparison between dry and humid condition. Despite the presence of water vapor, O<sup>2-</sup> species was involved in oxygen adsorption behavior for Fe-modified SnO<sub>2</sub> devices and it was similar to oxygen adsorption species in dry atmosphere. High response to H<sub>2</sub> was discussed in terms of decreasing donor density, protection effect and O<sup>2-</sup> adsorption species by catalytic effect of Fe ions on oxide surface. Consequently, surface modification by Fe/HF treatment seems to influence decreasing humidity dependence while keeping its gas sensing property in humid atmosphere.

Chapter 4 consists largely of results of DRIFTS analysis including single channel spectra, time-resolved spectra and multivariate curve resolution in order to clarify the relationship between surface chemistry with OH group and gas sensing behaviors. This analysis presented the possibility of easy access to confirm the gas sensing mechanism. Surface modification using 0.22 mol% Fe ions not only increases responses to H<sub>2</sub> in dry and humid air through receptor function of the additive, but also makes the water vapor effect smaller. The reason for this behavior is most probably related to the fact that surface modification with Fe ions creates new adsorption sites for OH groups though the kinetically separable component results of Fe-modified SnO<sub>2</sub> materials. As the concentration of Fe ions is increased, the new sites increase total amount of surface hydroxyls, that is, the formation of these new species might be the reason for the observed difference in the water effect. Hence surface modification of SnO<sub>2</sub> nanoparticles using Fe ions suggests an effective way to improve gas sensing performance and stability in humid atmosphere.

Final Chapter 5 is summarized the results of this study and presented the design of the surface modification using other metal ions as an outlook.