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論 文 要 旨

THESIS SUMMARY

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要旨（英文 800 語程度）
Thesis Summary (approx.800 English Words)

Gas sensors for machine olfaction are essential tools due to their wide range of applications in the industrial sector, food sector, health and environmental monitoring. Rapid advancements in information technology and embedded systems require these sensors to possess high sensitivity and selectivity for producing artificial olfactory systems where alternatives such as sensory test panels are costly, unreliable or time-consuming. For current gas sensors, although high sensitivity has been reported using various transduction principles, a limiting factor to further development has been in the discrimination of isomeric compounds or compounds with identical molecular weights. For isomer classification, novel sensing materials based on catalytic materials are explored in order to achieve sufficient selectivity.

For many years, catalytic materials have played a significant role in chemical analysis by modifying kinetics of target reactions. Thus, modern approaches in sensor technology have been to use these catalytic materials together with other sensing films to make novel nanocomposites. Such composites are easily applicable to electrochemical-type gas sensors where the sensing or working electrode can be modified by using electroanalytical techniques. Although traditional catalysts are of bulk size or nanoparticle size, recently single-atom catalysts or catalysts based on atomic size noble metals have been reported. By downsizing the catalyst from 'nano' to 'atomic' size, an increase in the catalyst's surface area contributes to sensitivity enhancement. For carbonaceous conducting polymers such as polyaniline, it has been shown that controlled growth of atomic size gold clusters are possible, which points to the possibility of preparing multiple sensing films with a wide range of selectivity.

Atomic-size catalysts are a new frontier in heterogeneous catalysis and in this research, atomic gold decorated on polyaniline conducting polymers are incorporated as nanocomposite films for electrochemical gas sensing. Atomic gold on polyaniline is a novel sensing film and its electrochemical sensing in gas phase is reported here for the first time. This study thus aims for the design and development of an electrochemical gas sensor with precisely defined atomic metal clusters on carbonaceous compounds as sensing films. Amperometric mode of electrochemical measurement is chosen as the transduction principle due to stable response and low cost. For electroanalytical measurements, cyclic voltammetry is used as the primary technique and the shape of cyclic voltammograms is studied to evaluate sensor response. Polyaniline polymerized on platinum macroelectrodes is subjected to a bottom-up electrochemical process whereby gold in its ion form is electrochemically decorated on metal binding sites created on polyaniline. Material characterization using scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDX) are reported and results indicate some key inferences.

For evaluation of electrochemical sensing behaviour of the nanocomposite, atomic gold clusters with different numbers of atoms (from 1 up to 4 atoms) have been prepared and their electrooxidation to propanol isomers in alkaline medium is reported. An odd-even pattern in catalytic activity with variation in atomic cluster size is observed. The pattern is similar to theoretically reported band gap energy variation of atomic gold clusters in related works. Due to its simplest structure but high catalytic activity, bi-atomic gold on polyaniline (PANI/Au_2) conducting polymer is chosen as a substrate for further investigation of sensor characteristics such as stability, reproducibility, detection limit and response time. The sensing film is also operable in gas phase and discrimination between gases of propanol isomers (normal-propanol and iso-propanol) based on features from their electrooxidation patterns are shown for the first time.

For fundamental analysis, the nanocomposite is also tested for other compounds such as linear alcohols (from ethanol up to pentanol) as well as aroma containing compounds such as geraniol (sweet rose smell) and benzyl alcohol (floral smell). While some compounds do not show sensor response, there are some compounds whose response can be quantified for specific applications. A study across functional groups for C3 compounds (compounds with 3 carbon atoms) is also reported. Most remarkable responses are obtained for alcohols and an indirect method for electrochemical sensing of ethyl formate based upon hydrolysis in alkaline ethyl formate is reported. Ethyl formate is a representative of the ester group and its scent is an indispensable component in the food industry, cosmetics industry and even as a fuel. For real-time gas sensing applications it is important to resolve mixtures of chemical signals which are otherwise non-additive in nature. Therefore in spite of a preliminary step in this study, binary mixtures of propanol isomers are roughly quantified using partial least squares regression. Finally, the miniaturization of the overall sensor from macroelectrode size to planar interdigitated microelectrodes is introduced. Due to their many advantageous properties, the use of alternative electrolytes such as room temperature ionic liquids (RTILs) are suggested.

Conducting polymers based on atomic metal catalysts are attractive new sensing materials for artificial olfactory systems. High sensitivity due to catalytic activity and modifiable selectivity due to precisely defined structure are expected using the proposed sensor.