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授与した学位	博　士
専攻分野の名称	学　術
学位授与番号	博甲第２８０８号
学位授与の日付	平成１６年　９月３０日
学位授与の要件	自然科学研究科物質分子科学専攻 （学位規則第４条第１項該当）
学位論文の題目	Studies on the Separation/Concentration and Determination of Trace Amounts of Gaseous Substances Using Flow-Based Analytical Methods （フロー分析法を用いる微量ガス状物質の分離・濃縮と定量に関する研究）
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学位論文内容の要旨

In recent decades, air quality has become an issue of great concern as more and more studies have shown. A number of studies have shown the great impact of atmospheric pollutant on environment and health. In most case trace gas analyses require on-site and continuous measurements because of fluctuations in concentration levels and difficulties with sample storage and/or the stability of stored samples. Moreover, regulated values of air pollutants in the atmosphere are continually lowed. Therefore, highly sensitive, rapid, simple and on-site monitoring of trace amount of air pollutants is urgently required.

In this study, the author aims at developing new flow injection (FI) techniques coupled with effective separation/concentration devices. A gas diffusion unit (GDU) was applied to the determination of citric acid based on the separation of gaseous ammonia followed by conductimetric detection. A newly designed GDU was applied for the determination of CO_2 in air by the development of a simple collection procedure with an appropriate absorbing solution. In this system, CO_2 in air was collected in a triethanolamine (TEA) solution as an absorbing solution with collection efficiency of almost 100%. A biporous hydrophobic PTFE device was developed for collection and concentration of gaseous pollutants, such as SO_2 and HCHO, in air by using a specific reaction for each analyte. Simple batchwise collection procedures for the determination of SO_2 and HCHO were also developed. The analytes in air samples were collected and concentrated in the TEA solution and the purified water for SO_2 and HCHO, respectively. Furthermore, portable flow injection analysis systems (PFA) were proposed for on-site monitoring of CO_2 and HCHO by incorporation with a batchwise collection procedure as described above. The proposed methods were successfully applied to real air samples and showed great potential for real time monitoring of pollutants in air.

In order to improve the sensitivity and to develop specific detection reagents for formaldehyde and ammonia detection, various kinds of reagents, were investigated: twelve kinds of β-diketones were tested. Using such reagents in the presence of ammonium acetate, formaldehyde was determined under moderate conditions, using the Hantzch reaction, followed by spectrophotometric and fluorometric detection. It was found that acetoacetanilide showed the possibility for formaldehyde determination. The key advantage of this reagent is its ability to react at room temperature and short reaction time.

論 文 審 査 結 果 の 要 旨

In recent decades, air quality has become a very important concern for human's health and sustainable environment; much more studies are required for accurate analysis of the atmosphere. Mostly, trace gas-pollutant analyses require on-site and continuous measurement, because of fluctuations in concentration levels and difficulties with sample storage and/or the stability of analytes in stored samples. Moreover, regulated values of air pollutants in the atmosphere are usually very low; therefore highly sensitive, rapid, simple and on-site monitoring of trace amounts of air pollutants has been urgently required.

In this study, the author aims at developing new flow-based techniques coupled with small-sized effective separation/concentration devices, such as a gas diffusion and a chromatomembrane device. The gas
diffusion unit (GDU) was applied to the gaseous ammonia separation, followed by conductimetric detection. Further, a newly designed GDU was developed, and applied to the determination of carbon dioxide (CO_2) in the atmosphere by coupling with a simple batchwise collection procedure using an appropriate absorbing solution and a plastic syringe. Using such a collection system, CO_2 in air was collected in a triethanolamine (TEA) solution as an absorbing solution with the collection efficiency of more than 97%.

A biporous hydrophobic PTFE device was developed for the collection/concentration of trace amounts of gaseous pollutants, such as SO_2 and HCHO, in air; the analyte concentrations in the absorbing solution were measured by using a specific reaction for each analyte. Simple batchwise collection procedures for the determination of SO_2 and HCHO were also developed. The analytes in air samples were collected and concentrated in the TEA solution and the purified water for SO_2 and HCHO, respectively.

For on-site analysis of the atmosphere, a portable flow injection analysis system (PFA) was proposed for the monitoring of CO_2 , SO_2 and HCHO by coupling with the simple batchwise collection/concentration procedure. The proposed methods were successfully applied to real air samples, which demonstrates that the methods will be potentially used for real-time rapid monitoring of air pollutants in the atmosphere.

In order to improve the sensitivity and to develop specific detection reagents for formaldehyde, as well as ammonia detection, various kinds of reagents were investigated: twelve kinds of β-diketones were tested. It was found that acetoacetanilide showed the good possibility for formaldehyde determination. Using this reagent in the presence of ammonium acetate, formaldehyde was found to react under moderate conditions, followed by a spectrophotometric and fluorometric detection. The main advantage of the proposed novel reagent is that the reaction can proceed at room temperature and within a short reaction time.

In view of original/creative contents and results in this study, the committee evaluated the dissertation as PhD degree's worth of research.