

[高度技術教育研究センター]

[区 分 A]

Complexsation of C₆₀ Fullerene with Cholesteryl Group-bearing Pullulan in Aqueous Medium

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Chem. Lett., 64-65 (2000)

Water-soluble complex between C₆₀ fullerene and cholesteryl group-bearing pullulan (CHP) was prepared. C₆₀ fullerene was dissolved in pyridine (10% v/v) in advance and then mixed with an aqueous CHP suspension (0.1 mg ml⁻¹). The particle size of the formed complexes varied from 60 nm to 150 nm by the concentration of aqueous pyridine in final solution. The complex could retain its integrity for a long period of time without destruction upon heating or freezing.

Utility of Liposomes Coated with Polysaccharide-bearing 1-Aminolactose as targeting chemotherapy for AH66 Hepatoma Cells.

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Onchology Reports, 7, 107-111 (2000)

The cell recognition element is very important for drug delivery systems. We synthesized cholesteryl pullulan (CHP) bearing 1-aminolactose (1-AL) and introduced a saccharide, cholesteryl pullulan bearing 1-aminolactose (1-AL / CHP), to an outer layer of the conventional liposome as a cell recognition element. Lectin recognized the α -galactose by aggregation of 1-AL / CHP coated liposome (1-AL / CHP liposome). The uptake of this liposome to AH66 rat hematoma cells was greater than in liposomes without 1-aminolactose *in vitro*. Furthermore, 1-AL / CHP liposomal adriamycin showed a stronger antitumor effect in comparison with other types of liposomal adriamycin *in vitro*. When *in vitro* tumor-targeting efficacy was investigated in AH66 tumor transplanted mice using ³H-liposome, the tumor / serum radioactivity ratio in mice injected with 1-AL / CHOP liposome was higher than that of mice injected with other liposomes. These observations suggest that 1-AL / CHP liposome is considered to be a good carrier of anticancer drugs for the active targeting of tumor cells.

Controlled Association of Amphiphilic Polymers in Water : Thermosensitive Nanoparticles formed by Self-assembly of Hydrophobically-Modified Pullulans and Poly(N -isopropylamides)

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Macromol., 33, 3244-3249 (2000)

Thermoresponsive hydrogel nanoparticles were prepared by self-assembly of two different

hydrophobically modified polymers, namely a cholesterol-bearing pullulan (CHP) and an copolymer of N-isopropylacrylamide (NIPAM) and N-[4-(1-pyrenyl)-butyl]-N-n-octadecylacrylamide] (PNIPAM-C₁₈Py). The interactions between CHP and PNIPAM-C₁₈Py were investigated by fluorescence spectroscopy, dynamic light scattering, and size exclusion chromatography. After ultrasonication of a mixture of CHP and PNIPAM-C₁₈Py (5:1 by weight) at 25 °C, monodisperse nanoparticles (Dh = 45 nm) were obtained, consisting of self-assembly of the two polymers associated via their hydrophobic moieties. Evidence from fluorescence and dynamic light scattering demonstrated that, above 32 °C, the lower critical solution temperature (LCST) of PNIPAM-C₁₈Py, the colloidal mixed nanoparticles increase in diameter (from 47 to 160 nm), but no macroscopic aggregation could be detected. This phenomenon was thermoreversible: upon cooling the particles recovered their original diameter.

Selective Uptake by Cancer Cells of Liposomes Coated with Polysaccharides Bearing 1-Aminolactose

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We investigated the selective uptake of liposomes chemically modified by polysaccharides-cholesterol derivatives with 1-aminolactose (lactose) in two human hepatoma cell lines (HuH7 and Alexander), a human colon cancer cell line (FCC) and a human lung cancer cell line (KNS). The uptakes of the labeled liposomes alone (conventional liposomes), those with cholesterol pullulan (CHP) and with lactose (lactose CHP) were compared in four cancer cells and normal rat hepatocytes after 3 hours of incubation. The radioactivities of the lactose CHP were 4.4, 4, 3.4 and 4.4 times greater than those of CHP in HuH7, Alexander, FCC and KNS cells, respectively, after 3 hours of incubation. All the above difference were statistically significant (p < 0.01). No statistically significant difference were seen in the case of hepatocytes. Thus, cancer cells have a common affinity with lactose CHP liposomes, however, these mechanisms appear to have no connection with the galactose-specific asialoglycoprotein receptors of hepatocytes.

Synthesis and Function of Sialic Acid-Conjugated Cholesterols as Ganglioside Analogs: Their Reconstitution to Liposomes and Interaction with Rat Lymphocytes.

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Proc. Japan Acad., 76 (B), 63-67 (2000).

Sialic acid-conjugated cholesterols (SC) with different spacer lengths were synthesized as ganglioside analogs. The interaction of SC (12 mol%)-containing liposomes with rat lymphocyte cells was investigated by monitoring increases in the intracellular calcium signal under a confocal fluorescence microscopy. Cells were stimulated strongly in the presence of SC-containing liposomes, while free SC not reconstituted to liposomes did not significantly stimulate the cells. The reconstitution of sialolipids to liposomes play a crucial role in the stimulation of cells. The efficiency of SC-containing liposomes was comparable to that of the naturally occurring gangliosides,

GT1b-containing liposomes.

HCl GAS SENSING PROPERTIES OF TPPH2 DISPERSE IN VARIOUS COPOLYMERS.

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Sensors and Actuators B, Vol. 65, pp138-140, (2000).

Composite films of tetraphenylporphyrin embedded in various of polymer matrix were prepared and their optical response to HCl gas was examined. The absorbance of the Soret and Q-bands for free-base tetraphenylporphyrin is reversibly sensitive to ppm levels of HCl. The lower Tg of polymer matrix than the sensing temperature is effective to enhance the sensitivity of the Soret- and Q-band region.

OPTOCHEMICAL HCl GAS DETECTION USING ALKOXY SUBSTITUTED TETRAPHENYLPORPHYRIN-POLYMER COMPOSITE FILMS. EFFECTS OF ALKOXY-CHAIN LENGTH ON SENSING CHARACTERISTICS.

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Sensors and Actuators B, Vol. 76, pp,42-46 (2001).

Composite films of 5,10,15,20-tetra (4'-alkoxyphenyl) porphyrin (TP(OR)PH₂) embedded in various of polymer matrices were prepared and their optical responses to HCl gas were examined. The absorbance of the Soret and Q-bands for free-base TP(OR)PH₂ is reversibly sensitive to sub-ppm levels of HCl. The lower Tg of polymer matrix than the sensing temperature is effective to the response and recovery behaviors. A high sensitivity to sub-ppm levels of HCl was achieved using the TP(OC₄H₉)PH₂-BuMA composite films.

Na₂O-RE₂O₃-SiO₂ (RE = Sm, Gd, Dy, Y, Ho, Er, Yb) 系ガラスの作製とその電気特性

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日本セラミックス協会学術論文誌 108[8] 774-776 (2000)

出発組成が 5Na₂O · RE₂O₃ · 8SiO₂ (RE = Sm, Gd, Dy, Y, Ho, Er, Yb) からなるガラスを作製し、そのナトリウムイオン導電性と希土類元素のイオン半径の関係についての検討を行った。作製したガラス試料の密度は、対応するセラミックス組成の理論密度と良く一致した。また、結晶化開始温度と結晶融解温度は、含有する希土類元素のイオン半径の増大とともに低下する傾向が見られた。200 における最も高い導伝率は Yb 系ガラスにおける 1.55 × 10⁻⁴ [S · cm⁻¹] で、希土類元素のイオン半径が増大するに従って、導伝率はわずかながら減少する傾向が見られた。

[区 分 B]

Biosimulation with Liposomes and Lipid Monolayers.

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"Physical Chemistry of Biological Interfaces" ed. By A. Baszkin and W. Norde, Marcel Dekker, Inc., New York, Chapter 10, 307-357 (2000).

Biosimulation, or mimicking of nature, refers to the simulation of the structure and function of any component of living systems composed of small molecules such as amino acids, lipids, nucleotides, saccharides, coenzymes, or hormones; biomacromolecules such as peptides, proteins, enzymes, DNAs, RNAs, or polysaccharides; and even tissues, organs or cells. Biosimulation involves astute use of the chemistry and physicochemistry of supramolecular assemblies of lipids, proteins, or polysaccharides. Although the primary purpose of biosimulation is, of course, to understand detailed mechanisms of living systems at the molecular level, the information and knowledge obtained through these basic investigation are beneficial in developing new biomaterials and biosystems designed for biotechnology and medicine.

[区 分 D]

天然由来化合物を基盤とする機能性超分子材料研究会

砂本順三*

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(財)新産業創造研究機構活動報告書

本研究会の本来の趣旨に従って、種々の角度から生物分解性天然由来高分子の材料としての機能開発研究に携わっている大学等の研究者に主としてシーズの提供を頂き、参加企業のニーズとのマッチングを探った。その結果種々のレベルではあるが、幾社かからの提案がえられ、次年度はより踏み込んだ、即ち研究開発の焦点を更に絞り込んだ形での研究会とすることが発案された。

移乗用車椅子と移乗補助車の開発

長田修次

新居浜工業高等専門学校機械工学科

ウエルフェアテクノシステム研究開発 (新居浜) 成果報告書 平成 12 年度 8 ~ 12 頁 (平成 12 年 3 月)

自分一人では体を移動させることが出来ない人を、ベッドから車椅子、車椅子から自動車などの双方向へ、移乗を容易にする車椅子および移乗補助車の開発を行った。本開発は、現状の車椅子の機能を全く損なわないで移乗用に改造した移乗用車椅子と、女性や高齢者でも 1 人で、要介護者を任意の所へ移動させることが出来る移乗補助車の 2 機構に分れるが、両機構の 1 号機の試作品を完成した。車椅子の乗り心地は現状と変わらない上、移乗時に要介護者を持ち上げる力も小さく、安心して移動できるので、搭乗者の昇降機構の電動化、軽量化を図り、実用化を目指したい。

移乗型簡易トイレの開発

長田修次

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ウエルフェアテクノシステム研究開発 (新居浜) 成果報告書 平成 12 年度 13 ~ 17 頁 (平成 12 年 3 月)

自分一人では体を移動させることが出来ない人を、女性や高齢者が 1 人でベッドからトイレへ容易に移

乗させる事ができる移乗型簡易トイレの開発を、移動台車に乗せた市販の簡易トイレに、本開発の移乗設備機構を取り付けて実現した。本品は上記目的を満たした上、 ベッド高さに合わせて調節可能、 使用中の背もたれ、肘おきの設置、 使用後は移乗設備機構を収納し、空き場所へ容易に移動でき、使い便利は良い等の特徴を有し、実用性は充分あると思われる。

〔 区 分 E 〕

超分子ナノサイズ微粒子の分散液構造と機能

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粉体工学会 2000 年度春期研究発表会 2000. 5

本講演では、水中における疎水化多糖の会合制御という観点から、疎水化プルラン(CHP)の分子量、疎水基の構造、また主鎖高分子と疎水基を結ぶスパーの有無等の違いにより、水溶液中で形成される微粒子集合体の会合特性を比較検討した結果について以下の順序で紹介したい。

水飴を原料とした生医用材料の特性

砂本順三*

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天然由来化合物を基盤とする機能性超分子材料研究会第 1 回講演会 2000. 5

数分子の高分子が会合し、相互に入り組んだ構造をした相互進入高分子はしばしば網目高分子ゲルを形成する。このようなゲルの持つ構造特性に新たな合目的性の機能を付与することが可能である。砂本らは天然由来多糖を部分的に疎水化し、それを水中に分散させることにより多糖の自己会合が起こり、構造の制御されたナノサイズのヒドロゲル微粒子の得られることを示してきた。具体的にはコレステロールやアルキル基を置換したプルラン水中で自発的に会合して、直径約 30nm のナノ微粒子を形成し、疎水基間の会合が確認できた。このヒドロゲル微粒子はナノテクノロジーとして極めて多才な機能を発現することが紹介された。

Specific Partition of Surface-Modified Liposomes in PEO and Polysaccharide Aqueous Two-Phase System

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13th International Symposium on Surfactants in Solution (SIS-2000) 2000. 6

(1) Hydrophobized polysaccharides such as cholesterol-bearing pullulan (CHP), dextran (CHD) and mannan (CHM) effectively coat the liposomal surface. To understand carbohydrate-carbohydrate interaction in water, partition of the hydrophobized polysaccharide-coated liposomes in an aqueous two-phase system (PEO (top)/pullulan (bottom) or PEO (top)/dextran (bottom)) was investigated. Conventional liposomes without polysaccharide coat mostly locate at the interface between the two polymer phases. The polysaccharide-coated liposomes were partly partitioned to the bottom polysaccharide rich phase. This specific partition depended on the structure of the hydrophobized polysaccharide on the liposomal surface. Affinity between the polysaccharide on the liposomal surface and that in the bulk bottom phase controls the efficiency of partition. (2) Partition of ganglioside (GM3, GD1a, GD1b or GT1b)-reconstituted liposomes was also investigated using the same strongly affected by the composition of the buffer employed. As decreasing the concentration of sodium phosphate, the partition of the negatively charged liposome largely increased in the bottom

dextran-rich phase. This partition to the dextran-rich phase depended also on the chemical structure of the saccharide moiety on the liposomal surface. The affinity of ganglioside being on the liposomal surface to dextran was the following sequence; GT1b > GD1a > GD1b > GM3. The partition of the liposomes to the top PEO-rich phase was negligibly small.

機能化リポソームを用いた新たなバイオテクノロジーの開花

砂本順三*

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天然由来化合物を基盤とする機能性超分子材料研究会第2回講演会 2000. 7

機能性脂質の合成と特性解析について紹介。融合性脂質、人工境界脂質および抗原性脂質などの各種機能を有する脂質の分子設計と合成。

Effective Reconstitution of Cell Membrane Proteins into Artificial Cell Liposome

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R&D Meeting of Japan Marine Science & Technology Center 2000. 8

In cell membranes, various proteins associate with the membranous lipids and play an important role in intercellular communication, cell-cell adhesion, signal transduction, mass transport across the cell membrane and so forth. Nevertheless, the study of membrane receptor proteins is much behind compared with that of soluble proteins. The perfect extraction of membrane proteins from intact cells and the subsequent reconstitution to a simpler artificial system without any denaturation and deactivation both are basic requirements in the investigation of membrane proteins. From this point of view, liposome seems to be a useful vehicle for the membrane proteins to be reconstituted, because the structural characteristics of liposomal membrane are similar to those of cytoplasm membrane.

Partition of Oligosaccharides in an Aqueous Two-Phase System

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International Symposium on Physical Basis of Adsorption, Okayama, Japan 2000.11

The partitions of linear oligosaccharides and cyclodextrins were investigated in three kinds of aqueous two-phase systems (PEO / dextran, PEO / pullulan and PEO / mannan) to obtain information about the effects of chemical structure of oligosaccharides on the partition of oligosaccharides and the oligosaccharide-polysaccharide interaction in water.

Cell Growth Activity of Synthesized Polyamionic Polymers

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Poly Millennium 2000, Hawaii, USA 2000. 12

A synthetic polyanionic polymer, poly(maleic acid-*alt*-7,12-dioxaspiro[5.6]dodec-9-ene) (MA-CDA), was found to enhance the growth of L929 and STO fibroblasts. After three days, at the optimum polymer concentration, the cell numbers increased 3.7-fold for STO cells and 1.9-fold for the L929 cells compared with the cells cultured in a comparable condition without MA-CDA. The cell growth promotion by MA-CDA required no other external growth factor present in a medium. In addition MA-CDA did not

induce the autocrine secretion of growth factors. These results indicate that MA-CDA has a direct growth promotion effect on these fibroblasts. This growth factor-like activity of MA-CDA is novel and unique among known synthetic polymers. Another polymer structurally related to MA-CDA, poly(maleic acid-alt-styrene), also showed the growth promotion activity similar to that of MA-CDA.

Polymers Secreted from Marine Microorganisms

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Macrofouling by marine organisms such as tunicates and barnacles is a serious problem for the shipping industry where foulers, attached to ship hulls, cause an increase in the fuel consumption. To prevent macrofouling, various coatings have been developed. However, the traditional antifouling coatings based on organic tin compounds and broadly toxic biocides have caused serious environmental pollution. Therefore, new methods for non-toxic control of marine biofouling have been of considerable interest.

Marine Technology Institute of Nippon Paint Co., Ltd. recently found the fouling-free natural film, Biojelly[®], that was produced by marine microorganism. This is a new method for macrofouling control which is required to be non-toxic and reproducible by marine microorganism.

In order to investigate the formation of Biojelly[®], we isolated several marine microorganisms on nutrient agar containing 2.4% NaCl from Biojelly[®]. Isolated microorganisms were cultivated at 30 °C in natural seawater or artificial seawater medium containing 0.1% yeast extract and 0.5% glucose. One of them, strain SHY1-1 produced polymeric film in the medium at 30 °C for 1 week in both shaking and static cultivation. The production of the film was improved by increasing the concentration of yeast extract and glucose. Immobilized strain SHY1-1 in nutrient agar plate also secreted the polymeric film on the agar surface in artificial seawater medium. The culture broth of strain SHY1-1 was very viscous, and white compounds were precipitated by adding 3 volumes of ethanol to the supernatant. These results indicate that strain SHY1-1 secreted a sort of polysaccharide to the culture supernatant. Strain SHY1-1 was allocated to the genus *Alteromonas* due to the morphological and physiological properties.

The relationship between the film and secreted polysaccharide is under investigation. We will also discuss the structure and biocidal function of these polymers.

A simple, small sized lock-in light detection system using a gated avalanche photodiode

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Optics Japan 2000 in KITAMI, 8aE21, p217-218, (2000)

By taking advantage to superiority in gain of the gate-biased avalanche photodiode (APD) over the conventional dc-biased APD, we have constructed a simple, small sized, lock-in light detection system. The APD is operated at a frequency of $2f$ ($= 20$ kHz) and then its output signal is fed into a compact, phase-sensitive detector IC which works at f ($= 10$ kHz). The system is effective for detecting weak light signal superimposed on a strong background light level.

OPTOCHEMICAL HCl GAS DETECTION USING ALKOXY SUBSTITUTED TETRAPHENYLPORPHYRIN-POLYMER COMPOSITE FILMS. EFFECTS OF ALKOXY-CHAIN LENGTH ON SENSING CHARACTERISTICS.

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第8回化学センサ国際会議 (スイス・バーゼル)、平成12年7月3日

Composite films of 5,10,15,20-tetra(4'-alkoxyphenyl)porphyrin(TP(OR)PH₂) embedded in various of polymer matrices were prepared and their optical responses to HCl gas were examined. The absorbance of the Soret and Q-bands for free-base TP(OR)PH₂ is reversibly sensitive to sub-ppm levels of HCl. The lower Tg of polymer matrix than the sensing temperature is effective to the response and recovery behaviors. A high sensitivity to sub-ppm levels of HCl was achieved using the TP(OC₄H₉)PH₂-BuMA composite films.

Keywords: gas sensor; optochemical sensor; UV-Vis spectrum; porphyrin; hydrogen chloride; environmental monitoring

OPTOCHEMICAL HCl GAS DETECTION USING mono-SUBSTITUTED TETRAPHENYLPORPHIN-POLYMER COMPOSITE FILMS

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第8回化学センサ国際会議 (スイス・バーゼル)、平成12年7月3日

Hydroxy / alkoxy substituted tetraphenylporphin were synthesized and their Soret- and Q- bands changes with HCl gas in ppm levels were examined. The both band was influenced by HCl gas concentration and their changes in a sub-ppm levels HCl were decreased with an increase in the alkoxy chain length. The changes of the absorbance of the Soret- and Q(0-0)-bands were enhanced by replacing ethylcellulose with poly-hexylmethacrylate as a matrix while the recovery times prolonged. The introductions of electron donating substituents to para-phenyl positions of the porphin ring result in the basicity of pyrrole nitrogens.

種々のアルコキシル基を含むテトラフェニルポルフィリン誘導体の合成およびそのオプティカルセンサ特性について()

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新居浜高専^{*1}、愛媛大工^{*2}

第31回複素環化学討論会 (北九州市小倉区)、平成12年10月3日

5,10,15,20-テトラフェニルポルフィリン(TPPH₂)のフェニル基のパラ位に鎖長の異なるアルコキシル基を導入した 5,10,15,20-テトラ(4'-アルコキシフェニル)ポルフィリン(TP(OR)PH₂)を合成し、各種スペクトルにより同定した。特に、それらのポルフィリンのNMR スペクトルにおいて、イソプロポキシル基のメチン水素が他のポルフィリンのアルコキシル基の酸素原子の 位の水素に比べて、0.8ppm 低磁場にシフトしていることが分かり、空間的にある位置に固定化されている興味深い知見が得られた。さらに、これらのポルフィリンの酸-塩基応答性は UV スペクトルの測定により確認し、これらのポルフィリンとポリマーの複合膜を作成し、HCl ガスに対するセンサ特性について検討した。

電子供与基を含むテトラフェニルポルフィリン誘導体の合成およびそのオプティカルセンサ特性について()

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第31回複素環化学討論会(北九州市小倉区)、平成12年10月3日

近年、NO_x、SO_x等の有害物質による環境汚染が問題となり、これらの規制対象物質のモニタリングシステムのための化学センサの開発が盛んに行われている。これまでに我々は、5,10,15,20-テトラフェニルポルフィリン(TPPH₂)をエチルセルロース等のポリマーに分散した複合膜を用いて HCl ガスオプティカルセンサの開発を行っている。その展開として、本研究では、TPPH₂のフェニル基のパラ位に電子供与性置換基として、水酸基や鎖長の異なるアルコキシ基を導入した種々の TPPH₂ 誘導体を合成し、ポリマー・マトリックスとして-72 から 125 までのガラス転移温度(T_g)であるビニルポリマーをラジカル重合により合成するとともに、HCl ガスに対するセンサの感度、応答および回復速度に及ぼす TPPH₂ 誘導体の置換基とポリマー・マトリックスの T_g による影響について検討した。

種々のアルコキシ基を含むテトラフェニルポルフィリン誘導体を用いたオプティカル HCl ガスセンサ素子の特性(2)

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日本化学会第79回春季年会(神戸市)、平成13年3月28日

環境汚染物質である HCl ガスをサブ-ppm レベルで検出するため本研究では、種々のアルコキシ基を5,10,15,20-テトラフェニルポルフィリン(TPPH₂)のフェニル基の2、3、あるいは4位に導入した TPPH₂ 誘導体を合成し、それらをポリマーマトリックスに分散させた複合膜と TPPH₂ を分散させた複合膜の HCl ガスに対する感度・応答速度等について比較検討したので報告する。

種々のヒドロキシアルキル基を含むテトラフェニルポルフィリン誘導体を用いたオプティカル HCl ガスセンサ素子の特性

朝比奈俊秀^{*1}、久保暁子^{*1}、中川克彦^{*1}、堤 主計^{*1}、田淵研三^{*1}、ヘルスプリヤノト^{*2}、青野宏通^{*2}、定岡芳彦^{*2}

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日本化学会第79回春季年会(神戸市)、平成13年3月28日

環境汚染物質である HCl ガスをサブ-ppm レベルで検出するため本研究では、種々の鎖長のヒドロキシアルキル基を導入した5,10,15,20-テトラフェニルポルフィリン誘導体を合成し、それらをポリマーマトリックスに分散させた複合膜と TPPH₂ を分散させた複合膜の HCl ガスに対する感度・応答速度等について比較検討すると、水酸基をフェニル基に直接導入したポルフィリンは応答速度及び感度の何れも優れているが、ベースラインへの復帰速度が他のポルフィリンに比べて遅くなる。

Na₂O-RE₂O₃-SiO₂ (RE:希土類元素)系ガラスの作製とその電気特性

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出発組成が5Na₂O・RE₂O₃・8SiO₂ (RE=La, Pr, Nd, Sm, Gd, Dy, Y, Ho, Er, Yb) からなるガラスを作製し、そのナトリウムイオン導電性と希土類元素のイオン半径の関係についての検討を行った。作製したガラス試料の密度はセラミックスの理論密度と良く一致した。

また、300 でのガラス試料の導電率は $10^{-4} \sim 10^{-3} \text{S} \cdot \text{cm}^{-1}$ で、Shannon らが報告した $\text{Na}_5\text{YbSi}_4\text{O}_{12}$ セラミックの導電率より一桁低い値であった。

〔 区 分 G 〕

車椅子収納リフター

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身体障害者や高齢者が使用している車椅子を乗用自動車に収納するためのリフターで、特別な動力を使わず、かつ安価な車椅子収納リフターを提供した。その原理は、スプリングによる伸縮力と、人による保持あるいは介添え程度の弱い力の補助により車椅子を吊り上げ、乗用車内へ車椅子を誘導したり、収納したり、引き出したりするものである。