〔 高度技術教育研究センター 〕

〔区　分　A〕

Partition of Linear and Cyclic Oligosaccharides in Aqueous Two-Phase Systems

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J. Bioactive and Compatible Polym., 14, 6-16 (1999)

In aqueous two-phase systems such as PEO／pullulan and PEO／dextran systems, partition of linear and cyclic oligosaccharides was investigated. The α1-4 linked linear oligosaccharides such as glucose, maltose, maltotriose, maltotetraose, maltopentaose, maltohexaose, and maltoheptaose more partitioned to the bottom polysaccharide-rich phase than to the top PEO-rich phase. The partition coefficient, *K* (the ratio of the concentration of oligosaccharide in the polysaccharide-rich phase to that in the PEO-rich phase), increased with an increase in the glucose unit of the oligosaccharide. The extent of the partition of the oligosaccharides was affected by the structure of polysaccharide employed in the two-phase system. Compared with the PEO／pullulan system, the PEO／dextran system showed the higher selectivity in the partition of the linear oligosaccharides. The structural characteristics of dextran with branched skeleton would be more suitable for the multipoint interaction with the linear oligosaccharides. On the other hand, more cyclodextrins (CD, cyclic oligosaccharides) were found in the PEO phase than in the polysaccharide phase. The sequence of partition coefficient of cyclodextrins to the PEO-rich top phase was the following; β-CD ＞ α-CD ＞ γ-CD. Regarding the more partition of β-CD to the PEO phase, the adequate incorporation of the PEO chain into the β-CD cavity might be responsible.

Bone marrow-derived dendritic cells incorporate and process hydrophobized polysaccharide／oncoprotein complex as antigen presenting cells

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Int. J. Oncol., 14, 695-701, (1999)

We have previously shown that a novel hydrophobized polysaccharide／oncoprotein complex vaccine can induce immune responses against the HER2／neu／c-erbB2 (HER2) expressing tumors. Bone marrow-derived dendritic cells (DCs), as antigen presenting cells (APCs), are the first candidates for presentation of tumor antigens. The aim of this study was to see whether DCs are able to elicit antigen specific host immune responses by stimulating the proliferation of T cells after exposure to cholesteryl group bearing pullulan (CHP) and HER2 protein complex. Vaccination by CHP-HER2 complex was as effective as cholesteryl group bearing mannan (CHM) and HER2 complex on which we reported previously. Immunization of mice with HER2 expressing CMS17HE tumor cells generated both CD^{4+} T cells and CD^{8+} T cells reactive with CHP-HER2 complex pretreated DCs. In addition, immunization with either CHP-HER2 complex or HER2 protein alone could also generates both CD^{4+} T cells and CD^{8+} T cells specifically reactive with CHP-HER2 complex pretreated DCs. The complete

rejection of tumors occurred when immunization with CHP-HER2 complex pretreated DCs was started 10 days after tumor inoculation. Therefore, bone marrow-derived DCs pretreated with hydrophobized polysaccharide／oncoprotein complex are a powerful tool for enhancing the effectiveness of oncoprotein for anti-tumor vaccination, opening new options for immune cell therapy.

Molecular Chaperone-like Activity of Hydrogel Nanoparticles of Hydrophobized Pullulan: Thermal Stabilization with Refolding of Carbonic Anhydrase B

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Bioconjugate Chem., 10, 321-324 (1999)

We have been studying the formation of hydrogel nanoparticles by the self-aggregation of hydrophobized polysaccharide and the effective complexation between these nanoparticles as a host and various globular soluble proteins as a guest. This paper describes a new finding that refolding of the heat-denatured enzyme effectively occurs with the nanoparticles and β-cyclodextrin according to a mechanism similar to that of a molecular chaperone. In particular, the irreversible aggregation of carbonic anhydrase B (CAB) upon heating was completely prevented by complexation between the heat-denatured enzyme and hydrogel nanoparticles formed by the self-aggregation of cholesteryl group-bearing pullulan (CHP). The complexed CAB was released by dissociation of the self-aggregate upon the addition of β-cyclodextrin. The released CAB refolded to the native form, and almost 100% recovery of the activity was achieved. The thermal stability of CAB was drastically improved by capture of the unfolded form which was then released to undergo refolding.

Cell Specificity of Macromolecular Assembly of Choresteryl and Galactoside Groups-Conjugated Pullulan

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J. Bioactive and Compatible Polym., 14, 195-211 (1999)

Galactose or lactose groups were additionally conjugated to cholesterol-bearing pullulan (CHP). The CHP derivatives so obtained formed monodisperse nanoparticles upon the self-aggregation in water. Nanoparticles of galactoside-conjugated CHP self-aggregate were specifically internalized by rat hepatocytes and HepG2 cells. Galactoside-bearing CHP-coated liposome or oil droplet of *O/W*-emulsion was also taken up by HepG2 cells. Tissue distribution of the nanoparticle of CHP self-aggregate changed dramatically with chemical conjugation of the galactoside moiety. Galactoside-bearing nanoparticles were specifically accumulated in the liver.

Gelation of Cholesterol-Bearing Pullulan by Surfactant and Its Rheology

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Colloids and Surfaces A: Physicochem. Eng. Aspects, 147, 203-211 (1999)

Gelation of cholesterol-bearing pullulan (CHP) with SDS in water was studied by rheological measurements. The apparent viscosity of the CHP (2% (w／w))／SDS mixture increased with an increase

in the SDS concentration up to 1% and then decreased after a maximum. In the presence of large amounts of SDS, the CHP self-aggregate certainly dissociated. With 3% (w／w) CHP, a macroscopic gel was formed by the addition of SDS above 0.5% (w／w). At higher concentrations of SDS (above 4.5% (w／w)), the gel changed to a sol. The mechanism of the gelation and the transition to the sol is related to the formation of mixed aggregates between the cholesteryl groups of CHP and SDS. Due to the strong association of the cholesteryl groups, large amounts of SDS were required to achieve the complete solubilization of cholesteryl groups.

Oscillatory shear measurements were carried out for the gel of the CHP／SDS mixture. In the low frequency region (＜0.1 Hz), G'' (loss modulus) showed a maximum, while G' (storage modulus) reached a plateau with an intersection of the G'' curve. This is a trend typical of a Maxwellian fluid. An extremely long relaxation time (20 s) was observed for the CHP／SDS gel at relatively low SDS concentrations. Such a long relaxation time would be ascribed to the strong association of the cholesteryl groups of CHP.

Enzymatic Synthesis and Characterization of Amphiphilic Block Copolymers of Poly(ethylene oxide) and amylose

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Macromol. Rapid Commun. 20, 112-115 (1999)

Amphiphilic A-B block copolymers, methoxypoly (ethylene oxide)-amylose copolymers (MPEO-amylose), were synthesized by an enzymatic reaction using potato phosphorylase from an MPEO (Mw＝5000)-maltopentaosylamine derivative as a primer and α-D-glucose-1-phosphate as a substrate. MPEO-amyloses with various molecular weights of amylose (degree of polymerization, DP＝26, 36, 73 and 112 glucosyl residues) were obtained. None of the MPEO-amyloses (5mg／ml) precipitated in water containing 10 vol.% DMSO even after 24 h, even though native amylose (DP＝74) aggregated and precipitated within 1 h under the same conditions. The MPEO-amyloses also dissolved in chloroform containing 2 vol.% DMSO and in toluene containing 2 vol.% DMSO. The MPEO-amyloses effectively complexed with iodine in water.

Effect of Macromolecular Assembly of Galactoside-Conjugated Polysaccharide on Galactose Oxidase Activity

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Macromolecular Chemistry and Physics, 200, 1386-1392 (1999).

Galactose (Gal) was conjugated to cholesterol-bearing pullulan, CHP-108-0.9 (CHP), with a suitable spacer. The CHP derivative, Gal(6)-24-CHP-108-0.9 (Gal-CHP), forms a monodisperse nanoparticle upon self-aggregation in water (RG＝17.0 nm). The reaction with -D-galactose oxidase was investigated with various self-assembly systems of Gal-CHP, such as nanoparticle of Gal-CHP self-aggregate, Gal-CHP-coated liposome and Gal-CHP-coated oil droplets of an O／W-emulsion. The galactoside moiety conjugated to the polysaccharide derivative showed a much lower Km-value than free Gal itself, though the kcat-value of Gal-CHP was found to be smaller than that of free Gal. For the galactoside-conjugated polymer, the binding affinity increases due to the self-assembly

of the polymer backbone. The content of the galactoside moieties in the nanoparticle dramatically affects the kinetic parameters of the enzymatic oxidation. Substrate affinity is higher with either the Gal-CHP-coated liposomes or the oil droplets than that with the nanoparticle of the Gal-CHP self-aggregate. An increase in the local concentration of the galactose moiety in the vicinity of the enzyme is considered to be an important factor in the enhancement of the enzyme activity for the multivalent substrate.

Self-Aggregate Nanoparticles of Cholesteryl and Galactoside Groups-Substituted Pullulan and Their Specific Binding to Galactose Specific Lectin, RCA120

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Macromolecular Chemistry and Physics, 200,1554-1560 (1999)

The hydrophobized polysaccharide cholesterol-bearing pullulan (CHP) forms hydrogel nanoparticles upon self-aggregation in water. To endow cell specificity to nanoparticles of CHP, galactosides were additionally substituted to CHP with various spacer lengths and degrees of substitution. The resulting CHP derivatives also formed monodisperse nanoparticles by self-aggregation in water. All self-aggregate nanoparticles strongly bound to the β-D-galactose-specific lectin, RCA120 (Ricinus communis agglutinin). The affinity of substituted galactosides to the lectin was quantitatively studied using a fluorescence polarization technique by competitive binding with 4-methylumbelliferyl β-D-galactopyranoside (MUG). This affinity strongly depended on its chemical structure, degree of substitution, and spacer length. The binding constant per galactose moiety drastically increased by substitution to the polymer backbone.

Asymmetric Distribution of Artificial Boundary Lipid as Analysed by NMR Deconvolution

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Chem. Lett., 205-206 (1999).

Asymmetric distribution of an artificial boundary lipid between the outer and inner leaflets of multilamellar vesicles (MLVs) was determined by the modified ^{1}H-NMR peak deconvolution technique. The asymmetric location of the lipid was significantly affected by the method of vesicle preparation. This suggests that a relatively high energy state of lipid bilayer is required for this lipid to function in bilayer.

Induction of Acetylcholinesterase Release from Erythrocytes in the presence of Liposomes

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J. Biochem, Tokyo, 125, 876-882 (1999)

When human erythrocytes are incubated with liposomes, the release of acetylcholinesterase (AChE) occurs following an induction period [Cook *et al*. (1980) *Biochemistry* 19, 4601-4607]. However, the mechanism of the induction has not been elucidated. We examined the relationships among the lipid transfer from liposomes to erythrocytes, the morphological change of erythrocytes, the

fluidity of the erythrocyte membrane and the start of AChE simultaneously after an induction period. The morphological index (MI) of erythrocytes was approximately 2.8 at the beginning of the release, regardless of the induction period. AChE was not released from the erythrocytes of index 2.8 even in the presence of liposomes if the MI remained at 2.8. Therefore, for the release, erythrocytes needed a further increase of the MI from 2.8. As the rate of lipid transfer increased, the induction period became shorter. No significant lipid releases from erythrocytes was detected during the induction period. The initiation of the AChE release was not simply affected by the change in the membrane fluidity of erythrocytes upon interaction with liposomes. These results first demonstrate that AChE release in to the shed-vesicle and liposome fractions is triggered by a further increase of the MI from 2.8, which is induced by lipid transfer from liposomes to erythrocytes.

Self-Assembled Hydrogel Nanoparticles Composed of Hydrophobized Polysaccharides and Hydrophobized Poly(N-isopropylacrylamides), (in Japanese)

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J. Network Polym., 20, 171-176 (1999).

Thermosensitive hydrogel nanoparticles were prepared by self-assembly of two different hydrophobically-modified polymers, namely a palmitoyl group-bearing pullulan (C16P) and N-[4-(1-pyrenyl)butyl-N-n-octadecylacrylamide] (PNIPAM-C18py). We have reported previously that hydrophobized polysaccharides formed self-assembled hydrogel nanoparticles, in which the associated domains of hydrophobic moieties were cross-linking points. In this study, the interaction between C16P and PNIPAM-C18py was investigated by fluorescence spectroscopy and dynamic light scattering. Mixing of the hydrophobic moieties was followed by fluorescence of pyrene. C16P and PNIPAM-C18py formed polymer networks in water at 25°C through the association of the hydrophobic moieties of the two hydophobized polymers. After ultrasonication of the mixture of C16P and PNIPAM-C18py (5:1 by weight) at 25°C, monodisperse nanoparticles (Dh=45 nm) were obtained. The PNIPAM chains within the nanoparticles retained their thermosensitivity. A study by fluorescence demonstrated that, above the lower critical solution temperature (LCST) of PNIPAM-C18py (35°C) the original hydrophobic domains were destroyed and a separate PNIPAM rich phase formed in the nanoparticle. This phenomenon was thermoreversible. It was observed also that phase separation induced aggregation of the nanoparticles resulting in the formation of aggregates approximately 100 nm in diameter. This mixing of two polymers via mixed hydrophobic domains represents a new preparation method of functional hydrogel nanoparticles.

OPTOCHEMICAL HCl GAS SENSOR BASED ON TETRAPHENYL-PORPHINE-POLYMER COMPOSITE FILMS
-Effects of Polymer Matrix on Sensing Characteristics-

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Electrochemistry, Vol. 67, pp431-437, (1999).

Spectral changes of tetraphenylporphine-polymer composite films were examined for detection of ppm levels of HCl gas. The $TPPH_2$ exists as a mixture of neutral form [N] and dicationic form [D], i.e $N + 2HCl_{film} \rightleftarrows [D]$, $K=[D]/[N][HCl]^2$ and the concentration of HCl in polymer matrix is related

to HCl_{gas} with [HClfilm]＝a[HClgas]n. The concentration of [D] can be written: [D] ＝ Ka^2 $[HCl_{gas}]^{2n}$(1 ＋Ka^2 $[HCl_{gas}]^{2n})^{-1}$. For the polymers with glass transition temperature which is lower than the working temperature (318 K), both Ka^2 and n value increased with an increase in the Tg and reached a maximum and then decreased. The inner rotation of the molecular chain and micro Brownian motion for the polymer matrix affects on the sensing characteristics／absorption spectral changes with HCl gas sorption.

OPTICAL HCl DETECTION USING COMPOSITE FILMS OF TETRAPHENYLPORPHINE-POLYACRYLATE

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Chemical Sensors, Vol. 16, Supplement A, pp54-56 (2000)

HCl sensors using Tetraphenylporhine-polymer composite films were prepared and the effects of the glass transition temperature (Tg) of the polymer in the films were examined. A higher sensitivity was obtained when Tg of the polymer is lower (290～310K) than operating temperature (318K) of the sensors. This improvement is ascribed to an increase in free volume. However, the sensitivity were reduced when Tg value of the polymer is lower than 290K.

〔区　分　D〕

超分子集合系による新しいドラッグデリバリーシステムの開発

砂本順三*、Barenholz**、 Yechezkel**
*新居浜工業高等専門学校、 **The Hebrew University, Department of Biochemistry
平成10年度科学研究費補助金（国際学術研究）研究成果報告書（10044152）

疾病の治療では，薬剤は必要な時に，必要量だけを，治療すべき病巣へのみ送達することが理想である．そのためには生体投与時の毒性と免疫原性の軽減，安全性，薬効の持続性，保存時の安定性，製剤の容易性などのあらゆる要求に応えねばならない。それゆえ安全な生物分解性を示し，かつ細胞特異性に優れた適切なキャリアーと真薬とを超分子集合体として製剤化することが望ましい。この要望に応えるために，本研究代表者（砂本順三）と分担者（Yechekel Barenholz）は，共同で研究することを計画した。これまで研究代表者は天然由来多糖類を用い，一方分担者はリポソームを用いてそれぞれ独立に当該研究課題の分野で永年研究を展開しており，いずれも優れた成果を挙げ，国際的に高い評価をえてきた。本研究では（１）細胞特異性の疎水化多糖類の合成とそのin vitroテスト並びに（２）細胞特異性の疎水化多糖被覆リポソームに内包した薬剤の生体内組織分布，安全性と薬効の評価を計画した。計画実行に際しては，平成10年10月にBarenholz教授が来日し，平成11年３月に砂本教授が大学院博士課程の学生２名と共にイスラエルを訪問，研究討議と実験に携わった。検討課題１では，まづ新たにガラクトースおよびラクトース残基をコレステロール修飾プルラン(CHP)に置換することに成功した。このものおよびこのもので被覆したリポソームのいずれもが，ガラクトースレセプターをもつ肝実質細胞および HepG2 に特異的に取り込まれることが判明した。このものを更に FITC で蛍光ラベルして行った検討課題２ではラットへの静注時に肝臓への集積は定性的に認められたものの，残念ながら組織分布の定量的評価と免疫活性の評価は完結できず，今後の検討課題を残した。

横移乗車椅子の開発

長田修次*1、吉川貴士*1、桂　誠司*2
*1新居浜工業高等専門学校機械工学科，高度技術教育研究センター、 *2(株)タイワ

ウェルフェアテクノシステム研究開発（新居浜）
平成11年度　23頁　平成12年3月
　要介護者を介助者１人で容易にベットから車椅子、あるいはその逆方向に移せる車椅子を開発した。

車椅子収納リフターの開発
長田修次*[1]、谷口佳文*[1]、桂　誠司*[2]
　*[1]新居浜工業高等専門学校機械工学科，　高度技術教育研究センター、　*[2](株)タイワ
ウェルフェアテクノシステム研究開発（新居浜）
平成11年度　34頁　平成12年3月
　乗用車のトランク内へ車椅子を容易に収納させる補助リフターを開発した。

平成 10 年度　地域コンソーシアム研究開発事業「 ベンチャー企業育成型地域コンソーシアム（中小企業想像基盤型）」
「半導体プロセス排気ガスのプラズマ処理システムの開発」成果報告書
石川年明、出口幹雄
新居浜工業高等専門学校高度技術教育研究センター
　大気圧プラズマを用いた排ガス処理方式の要素技術について研究を行い、乾式処理方式については、バリア放電型とトーチ型について比較検討した結果、トーチ型の方が小型で分解率も高いことが判明した。湿式処理方式については、分解率100%を達成することができた。

〔区　分　E〕

Liposomal Simulation of Functions on Cellular Surfaces
J. Sunamoto
Niihama National College of Technology
The 1999 International Congress on Membranes & Menbran Processes, Tronto, Canada
1999.6
　Liposome is considered as a beneficial cell membrane model. However, we have to overcome many problems when we employ liposome as the useful material in biotechnology and medicine. For this purpose, we have developed several functional lipids and reconstituted in conventional liposome to enhance the function of liposome; namely, (1) artificial boundary lipid, (2) fusogenic lipid, and (3) antigenic lipid. In this paper, I would like to give an overview of these results obtained in our own laboratory.

Supramolecular assemblies for potent vaccine delivery
J. Sunamoto
Niihama National College of Technology
The Liposome Advances: Progress in Drug and Vaccine Delivery, London, UK
1999.12
　Supramolecular assembly is defined as self-organisation of molecules based on weak and noncovalent bonding such as hydrophobic association, hydrogen bonding, dipole-dipole interaction, ionic bonding and／or charge transfer interaction. When we design a potent vaccine delivery system, a totally noncovalent assembly upon self-organisation (*supramolecular assembly*) is proposed, in which true antigen and its carrier specific to immune responsive cells are noncovalently assembled. In this

case, however, we have to overcome several disadvantages of biochemical, chemical and physicochemical instability of the vaccine during the processes of formulation and the storing as well as after in vivo administration. Since 1989, we have been extensively investigating potent vaccine delivery using supramolecular assembly systems such as liposomes and self-associating polymers. In this paper, I would like to introduce several examples: (I) Priming for in vitro and in vivo anti-HTLV-1 cellular immunity with a specially designed liposome. (Ⅱ) Induction of in vitro and in vivo anti-tumour responses by sensitisation of mice with a liposome which bears tumour surface antigenic protein (TSAP). (Ⅲ) In vitro and in vivo cellular and humoral immune responses against HER2-expressing murine sarcomas using polysaccharide nanoparticle as complexed with oncoprotein. (IV) Effective immunisation of mice with bone marrow-derived dendritic cells which are pre-treated by polyssacharide／HER2 complex.

細胞膜の構造・機能特性をリポソームでシュミレーションする

砂本順三
新居浜工業高等専門学校
日本膜学会第 21 年会　1999.5

衆知の如く、リポソームは細胞の最も単純なモデルとしてデヴユーした。それ故、リポソームによる細胞膜のシミュレーションは当然過ぎる程の期待である。しかし、実際の細胞のあまりにも複雑な構造・機能を再現させるには通常のリポソームはこれまたあまりにも単純すぎる。そこで、単純なリポソームに何らかの修飾を施し、細胞のもつ極めて高度な機能に一歩でも近付いてその本質に迫り、また新しいシステムや材料の開発を目指すのも一つのアプローチである。演者はこれまで細胞膜の構造・機能特性を目的を定めて機能化したリポソームを用いてシミュレーションし、得られた情報を基にして細胞膜研究のための新しい技法の提案と医療やバイオテクノロジーでの応用展開を心掛けてきた。今回はそれらの内の幾つかの例を紹介したい。

超分子集合体を用いてのバイオシュミレーション

砂本順三
新居浜工業高等専門学校
第 14 回生体機能関連化学シンポジウム　1999.9

細胞膜上でのいくつかの現象とそれを動かしている脂質群の構造と機能を人工系でシュミレーションした。本講演では特に機能性脂質とその超分子集合系を中心にした以下のトピックスを紹介する。１．細胞表層での糖鎖の認識機能シュミレーション。２． 生体系での特異的結合における multivalency effect のシュミレーション。３． 細胞膜上での膜タンパク質の保持シュミレーション。４． 細胞融合のシュミレーション。５． 免疫原性を有する脂質の抗原性発現のシュミレーション。

大気圧プラズマによるＣＦ４の分解

石川年明、出口幹雄、メバルキ・ベンチェルキ、板谷良平
新居浜工業高等専門学校高度技術教育研究センター
第 17 回プラズマプロセシング研究会　2000 年１月

大気圧プラズマを用いたＣＦ４ガス処理方式の要素技術について研究を行い、乾式処理方式については、バリア放電型とトーチ型について比較検討した結果、トーチ型の方が小型で分解率も高いことが判明した。湿式処理方式については、分解率 100%を達成することができた。

大気圧ドライプラズマによるＣＦ４の分解

出口幹雄、石川年明、メバルキ・ベンチェルキ、板谷良平
新居浜工業高等専門学校高度技術教育研究センター
第 17 回プラズマプロセシング研究会　2000 年 1 月
　バリア放電型とトーチ型の大気圧プラズマについてＣＦ４の分解特性を比較検討した結果、トーチ型の方が小型で分解率も高いことが判明した。

大気圧ウェットプラズマによるＣＦ４の分解

石川年明、出口幹雄、メバルキ・ベンチェルキ、板谷良平
新居浜工業高等専門学校高度技術教育研究センター
第 17 回プラズマプロセシング研究会　2000 年 1 月
　湿式の大気圧プラズマを用いたＣＦ４ガス処理方式について研究を行い、分解率 100%を達成することができた。

テトラフェニルポルフィリン-アクリル酸エステルポリマー複合膜を用いた光学的　HCl ガスセンサ

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化学センサ研究会 (名古屋大学)、平成 12 年 3 月 31 日
　１．ガラス転移温度が測定温度よりもやや低いポリマーマトリックスを用いることで応答、回復性は良くなる。これは膜中の自由体積の増大により HCl ガスが拡散しやすくなったためと考えられる。
　２．測定温度より極めてガラス転移温度が低いポリマーマトリックスを使った場合ピークに割れやシフトが生じ、回復時間が遅くなる。これを改善するためには色素濃度を小さくするのが有効である。

Na_2O-RE_2O_3-SiO_2（RE：希土類元素）系ガラスの作製とその電気特性

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　Na_2CO_3、RE_2O_3及びSiO_2の混合物を溶融することによって、７種類のナトリウム－希土類－珪酸塩ガラス $(Na_2O)_{35.7}(RE_2O_3)_{7.2}(SiO_2)_{57.1}$ (RE＝Y、Sm、Gd、Dy、Ho、Er、Yb) を調製し、それらの電気特性について検討を行った。得られたガラスの密度は、$Na_5RESi_4O_{12}$ セラミックスの理論密度とよく一致していた。　また、結晶化温度は希土類元素 (RE) のイオン半径が大きくなるに従い低くなった。最も高い導電率 (200℃で $1.55x10^{-4}$ S cm^{-1})は、$(Na_2O)_{35.7}(Yb_2O_3)_{7.2}(SiO_2)_{57.1}$ で得られた。また、導電率は、希土類元素 (RE) のイオン半径が大きくなるに従い僅かながら低くなった。