〔 高度技術教育研究センター 〕

〔区　分　A〕

中山　享

Improvement of low temperature carbon combustion catalyst characteristic caused by mixing Bi_2O_3 with Tl_2O_3

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This study investigated the addition of various oxides to further improve the catalytic characteristics of Tl_2O_3, which offers a high carbon combustion catalytic capacity to lower the carbon combustion temperature of 660 °C by ~300 °C. Mixtures of carbon (2 wt%) with composite catalysts comprising 20 wt% Tl_2O_3-80wt% added oxide were analyzed using DSC. Bi_2O_3 offered the best improvement, where the exothermic peak temperatures for carbon combustion of carbon with various Tl_2O_3-x wt% Bi_2O_3 composites were lower than that of carbon with pure Tl_2O_3. Isothermal TG measurements were performed using a mixture of carbon and the Tl_2O_3–95 wt% Bi_2O_3 composite catalyst, where a 2 wt% weight loss (i.e. removal of all carbon) was achieved above 230 °C. A porous alumina filter was coated with the composite catalyst and carbon was deposited on the filter surface. The filter was held at constant temperatures under air flow, which confirmed that carbon was completely removed at 230 °C. This study demonstrated the potential for using these composite catalysts in self-cleaning particulate filters to decompose and eliminate fine particulate matter and diesel particulate matter generated from steelworks, thermal power plants, and diesel vehicles simply using the heat of the exhaust gas in a factory flue-gas stack or vehicle muffler.

中山　享

Lithium and sodium ion-exchange behavior of $(H_3O)Zr_2(PO_4)_3$ with three-dimensional network structure

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The ion-exchange behavior of H_3O^+ in $(H_3O)Zr_2(PO_4)_3$ and alkali metal ions, M^{I+}, was examined in an aqueous solution. The pH titration was performed using a 0.1 mol·L^{-1}–M^IOH solution (M^I=Li, Na, K, Rb, and Cs), while stirring and distributing $(H_3O)Zr_2(PO_4)_3$ in deionized water. In K, Rb, and Cs, the pH value increased to ten or more after adding the M^IOH solution. Therefore, it was believed that ionic exchange with H_3O^+ in $(H_3O)Zr_2(PO_4)_3$ did not advance. Conversely, ionic

exchange with H_3O^+ was observed at Li and Na. In Li only or Na only, Li was easily immobilized and leached, and Na was inferior to Li. In Li+Na, the immobilized amount of Li decreased, Li leaching was difficult, and the immobilized amount of Na increased.

中山　享

Elementary approach on the prediction of next material composition using AI technology: Improvement of characteristic by changing two components

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This study aims to identify the factors affecting the characteristics of samples, such as photoluminescence intensities, and identify the relationship between performance improvement and the search parameters for material composition. Subsequently, we optimize the experimental conditions to provide the maximum characteristic value. First, the process parameters are introduced as input values to the artificial intelligence (AI)-based model; then, we obtain a generalized equation to establish relationship between the characteristics of the samples and the process parameters. Subsequently, the new samples suitable for determining an accurate model and optimizing the process parameters are calculated and recommended to the user. Finally, the obtained formula is optimized, and the optimum values for achieving maximum characteristic are determined. Experimental validation using the AI program developed in this study found that the two components (x, y) that provide the strongest PL intensity in the $Sr_x(La_{10-x-y}Eu_y)(SiO_4)_6O_{3-x/2}$ (x=2-6, y=0.6-1.2) red-emitting phosphors can be easily estimated from approximately 10 initial data points.

中山　享

Crystal phase, electrical properties, and solid oxide fuel cell electrolyte application of scandia-stabilized zirconia doped with rare earth elements

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ZrO_2-10 mol% Sc_2O_3 (10ScSZ) has attracted attention as an electrolyte material for solid oxide fuel cells owing to its high conductivity. However, the phase transition between cubic and rhombohedral occurs in the range 500-600 °C, resulting in its rapid decrease in conductivity below 500° C. In this study, we determined the elements that can eliminate phase transition in the range 500-600 °C for 10ScSZ to realize high conductivity for all temperature regions. Rare elements were incorporated to 10ScSZ. X-ray diffraction and conductivity measurements were used to confirm the occurrence of phase transition. The results noted that the addition of La, Ce, Pr, Nd, Sm, Eu, Gd, and Tb improved the conductivity of 10ScSZ below 500° C, unlike that with the addition of Dy, Y, Ho, Er, Tm, Yb, Lu, and Sc. Further, although the addition of Ce to 10ScSZ was considered a solution, Nd and Sm were also noted to be effective additives.

中山　享

Effects of preparation molar ratio on immobilization of Cs and Sr in $(H_3O)Zr_2(PO_4)_3$ through heating in an aqueous solution

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We investigated the effects of the preparation molar ratios $(Cs/(H_3O)Zr_2(PO_4)_3$ and $Sr/(H_3O)Zr_2(PO_4)_3)$ on the immobilization of Cs^+ and Sr^{2+} ions in $(H_3O)Zr_2(PO_4)_3$ through heating at 250 ° C for 24 h in an aqueous solution using an autoclave. When the preparation molar ratios were 0.4 or lower for Cs and 0.2 or lower for Sr, the immobilization of the corresponding ions occurred almost to completion. On the other hand, for the molar ratios (1.0 for the Cs system and 0.5 for the Sr system) at which all the H_3O^+ ions in $(H_3O)Zr_2(PO_4)_3$ could be replaced by the Cs^+ or Sr^{2+} ions, the immobilization reaction did not proceed readily.

中山　享

PM2.5の燃焼触媒としての酸化タリウムの反応機構に関する考察

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優れた炭素燃焼特性を有する Tl_2O_3 に 7 種類の酸化物イオン伝導セラミックス $(ZrO_2)_{0.92}(Y_2O_3)_{0.08}$、$(CeO_2)_{0.8}(Gd_2O_3)_{0.2}$ 、 $La_2Zr_2O_7$ 、 $(Bi_2O_3)_{0.75}(Y_2O_3)_{0.25}$ 、 $(ZrO_2)_{0.90}(Sc_2O_3)_{0.10}$ 、 $La_{9.7}Si_6O_{26.55}$ 、 $La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.115}Co_{0.085}O_3$ を複合化し、炭素燃焼特性への影響を調べた。酸化物イオン伝導セラミックスとの複合化による Tl_2O_3 の炭素燃焼特性の向上は認められなかったが、x wt%$-Tl_2O_3+(Bi_2O_3)_{0.75}(Y_2O_3)_{0.25}$ (x = 5, 10, 20)複合体では Tl_2O_3 単独の炭素燃焼特性を維持していた。Tl_2O_3＋酸化物イオン伝導セラミックス複合体の炭素燃焼特性は、イオン伝導以外の機構が示唆された。

中山　享

Relationship between SiO_2 content and electrical properties of 8-mol% Y_2O_3-stabilized ZrO_2 electrolyte

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After preparing 8-mol% Y_2O_3-stabilized ZrO_2 electrolytes with SiO_2 contents to <40, 550, 1200, and 2800 ppm and attaching Pt electrodes, a direct-current (DC) load test wherein 1 V was applied at 800 ℃ in air atmosphere for 1000 h was conducted to investigate the effect of SiO_2 on the electrical properties. The DC resistance at 800 ° C for all samples increased immediately after the test was started, but tended to stabilize over time, and remained almost constant after approximately 600 h. The bulk and the grain-boundary resistances measured by the AC

complex impedance analysis at 300 °C after the DC voltage load test were increased in all samples compared to that before the test, but no clear correlation was found between the amount of SiO_2 and the resistance. Moreover, the electrode interface resistance measured by complex impedance analysis at 800 °C also increased after the DC voltage load test.

中山　享

Simple hydrogen gas production method using waste silicon

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　We investigated a simple and safe method for producing hydrogen using Si powder, which is discarded in the semiconductor industry. Using the reaction of generating hydrogen from Si powder and an aqueous NaOH solution, a simple hydrogen generator that imitated Kipp's apparatus was produced. Then, by combining this apparatus with a polymer electrolyte fuel cell, an automatic hydrogen generation system based on the amount of electric power required was proposed. Furthermore, it was found that hydrogen can also be generated using non-poisonous and deleterious substances $Ca(OH)_2$ and Na_2CO_3 instead of the deleterious substance NaOH and adding water to the mixture with Si powder. The by-products Na_2SiO_3 and $CaCO_3$ can be used as raw materials for glass. The simple hydrogen generator produced in this study can be used as a fuel supply source for small-scale power generation systems as an auxiliary power source.

中山　享

Tl_2O_3の低温炭素燃焼特性およびセルフクリーニング型 PM 燃焼分解除去フィルターへの応用

中山享*

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　単独酸化物 Tl_2O_3 を炭素燃焼触媒として用いた場合、炭素のみでの燃焼温度660℃が飛躍的に下がり300℃付近で炭素燃焼が可能であることを発見した。Tl_2O_3 および $Tl_2O_3＋Bi_2O_3$ 複合体の炭素燃焼特性、および $Tl_2O_3＋Bi_2O_3$ 複合体を低温炭素燃焼触媒としてコーティングした多孔質セラミックスからなる「PM 燃焼分解除去フィルター」を紹介する。$Tl_2O_3－Bi_2O_3$ 系低温炭素燃焼触媒をコーティングしたセラミックス多孔体からなるフィルターを工場の煙路や車のマフラーに配置することで排ガス中の PM2.5 や DEP を捕集し、排ガスの熱によって分解除去できる「自己クリーニング型パティキュレーションフィルター」の実現が期待できる。

堤　主計

An environmentally adaptable stereocomplex derived from lactide copolymers with improved UV shielding characteristics based on morphological changes

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This work attempted to fabricate biodegradable copolyester films that gradually adopt UV shielding characteristics based on changes in crystallinity resulting from the effects of temperature and humidity. The goal was to develop a novel UV shielding material capable of actively regulating the passage of UV radiation solely due to morphological changes rather than the addition of compounds such as TiO_2 or Fe_2O_3. The other aim was to obtain a polymer that would undergo hydrolysis after use to allow ready disposal. A biodegradable polymer with improved thermal and mechanical properties was fabricated by preparing a series of stereocomplex (St) specimens from optically active polymers. Films of these specimens were evaluated over time to assess UV transmittance, crystallinity and molecular structure while aging at 35 °C and a relative humidity of 85%. A series of St samples was made by combining poly(L-lactide(LA)-*ran*-trimethylene carbonate (TMC)) and poly(D-LA-*ran*-TMC) copolymers having various LA/TMC molar ratios. St specimens made with ratios of 91/9 and 92/8 in the two copolymers or both having an 88/12 ratio showed higher crystallinity than the original random copolymers. The crystallization of each St gradually increased during 20 weeks of storage at 35 °C and 85% relative humidity with simultaneous reductions in UV transmittance. The St composed of poly(L-LA-*ran*-TMC) (80/20) and poly(D-LA-*ran*-TMC) (81/19) was less crystalline and so it crystallized more rapidly during storage, similar to the original random copolymers. The St films composed of the random copolymers degraded more slowly than poly(L-LA-*ran*-TMC) with proteinase K.

〔区　分　C〕

中山　享
アパタイト型イオン伝導体の積層電解質を用いた CO_2 ガスセンサの応答特性

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固体電気化学セル(-)CO_2,O_2,Pt│$LiLaSiO_4$ / $La_{10}Si_6O_{27}$│Au、Li_2CO_3、CO_2、O_2(+)を用いて、空気中の二酸化炭素ガスセンサについて調べた。 電解質には、Li^+イオン伝導体とO^{2-}イオン伝導体を熱処理して作製した積層型イオン伝導体を使用した。 450～600℃の範囲では、センサ起電力 EMF は、ネルンストの法則に従って、CO_2分圧の対数の増加に伴って直線的に増加しました。一方、EMF は O_2 分圧の変化による影響をほとんど受けなかった。検出極では CO_2 による 2 電子反応と O_2 による 4 電子反応が起こっていた。加えて、 O_2 による 4 電子反応は検出極と対極の両方で起こっていた。この結果より、本センサが CO_2 に対して優れた選択性を持っていることがわかった。

中山　享
市之川鉱山産・輝安鉱の形態観察および元素分析

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用化学科

　愛媛県西条市の旧「市之川鉱山」から産出した輝安鉱について、エネルギー分散型分析装置付き電子顕微鏡を用いて形態観察および元素分析を、さらに波長分散型蛍光 X 線分析装置を用いて微量含有元素分析を行った。

中山　享
コマツナの発芽と生長へ及ぼすアルカリ金属の影響

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　$2.0 \times 10^{-5} \sim 1.0 \times 10^{-1}$ mol・L^{-1} の濃度範囲において 11 種類の異なる濃度の各アルカリ金属（Li、Na、K、Rb、Cs）を含有した試験用水を準備し、コマツナの発芽率と生長度合いを対照区（イオン交換水）と比較した。元素の違いによる発芽率の大きな差はみられなかったが、生長度合いについては Na の 1.6×10^{-3} mol・L^{-1} の濃度で、K の 8.0×10^{-4} と 1.6×10^{-3} mol・L^{-1} の濃度で対照区に対して 10〜12%の有意性がみられた。

堤　主計
L−ラクチドとトリメチレンカーボネートの開環共重合

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　本研究室では、ポリ乳酸の改質のために原料である L−ラクチド(L−LA) と共重合させるモノマーとして、δ−バレロラクトン（VL）、ε−カプロラクトン（CL）、1,5−ジオキセパン−2−オン（DXO）、テトラメチレンカーボネート（TEMC）などを用いて共重合体を合成し、分解性のほかに熱的特性や機械的特性などの物性を評価してきた。さらに、高収率かつ高分子量のポリ乳酸やそれらの共重合体を合成するために触媒として従来のオクチル酸スズを用いた重合方法の検討を行い、簡便な合成方法を確立している。本研究は、すでに確立した共重合体の合成方法を参考にトリメチレンカーボネート（TMC）を用いてポリ乳酸共重合体を合成し、その分子量および収率について比較検討を行った。

〔区　分　E〕

堤　主計
ステレオコンプレックス体形成によるポリ乳酸共重合体の物性変化

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　これまでに徐放剤の作製のためにポリ乳酸共重合体を用い、薬剤の含浸を行ってきた。薬剤を含浸させるために共重合体を可塑化させるがベースとなるポリマーの熱的特性が大きく影響する。徐放剤として利用するためには、熱的特性のほかに分解性や機械的特性も重要な要因となり、これまでに合成した共重合

体への薬剤含浸性や作製した徐放剤からの薬剤徐放性を検討してきた。これまでのポリ乳酸共重合体は主にランダム体として合成や各種物性を検討してきたが、今回はステレオコンプレックス体（SC 体）の作製とそれらの各種物性について評価した。ポリ乳酸の原料である L–ラクチド (L–LA) とトリメチレンカーボネート (TMC) の L 体のランダム共重合体と、同様に D–ラクチド (D–LA) を用いた D 体のランダム共重合体をそれぞれ合成し、これらの複合体である SC 体を作製した。共重合体や SC 体をフィルムにした後、DSC、XRD、FT–IR などにより物性値を測定・検討した。