公開講演会　最新化学談話シリーズ

平成 28 年度第 6 回談話会

ゼオライトによる CO_2 及び各種有害物質の吸蔵捕集

Capture of CO_2 and Other Harmful Species with Zeolites

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主催　九州大学理学部化学教室談話会

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近年、CO_2ガス吸蔵物質については広く研究されてきたが、その多くの多孔性物質がCO_2吸蔵捕集時に共存する水蒸気の吸着によって劣化・分解する問題を抱えてきた。本講演で紹介するゼオライト化合物ではCO_2とH_2Oの吸着サイトが高選択的に異なるサイトで起こるため、通常は水の吸蔵によってCO_2ガス吸蔵が阻害されるのに対し、このゼオライト化合物では、水蒸気共存下においてもCO_2の吸蔵捕集特性が全く阻害されないことはなく、かつ、水蒸気雰囲気下でも極めて安定であるという優れた特性が達成された(***Science* 2015**, *350*, 302-306)。また、放射性$^{137}Cs^+$除去用の多孔性物質としてのミクロ多孔性混合原子価バナドシリケートが優れた$^{137}Cs^+$除去特性を示すこと、並びに、CS^+に対する１８配位の特異構造を形成することを紹介する（***Angew. Chem. Int. Ed.* 2014**, *53*, 7203–7208）。さらに、原子炉炉心上層の気相部には水蒸気を多く含む酸性ガスに加え、各種の放射性ガス成分が発生する。主として、I_2及び有機ヨウ素が発生する。発表では、疎水性の全シリカゼオライト（silicate-1）が５Mの硝酸水溶液中でも安定であり、かつ、I_2、CH_3I、CH_3CH_2Iを高選択的に捕集する機能を持つことについて示す。その吸着特性は活性炭を凌ぐ点で注目される。これら物質がI_2を吸蔵した際に示す固体伝導物性についても興味深い知見が得られており、それについて紹介する（***Energy Environ. Sci.* 2015**, *9,* 1050-1062)。

（注）この講演は大学院集中講義の一部です。

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Capture of CO_2 and Other Harmful Species with Zeolites

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Capturing CO_2 from humid flue gases and atmosphere with porous materials remains costly because prior dehydration of the gases is required. A large number of microporous materials with physical adsorption capacity have been developed as CO_2-capturing materials. However, most of them suffer from CO_2 sorption capacity reduction or structure decomposition that is caused by co-adsorbed H_2O when exposed to humid flue gases and atmosphere. A highly stable microporous coppersilicate which has H_2O-specific and CO_2-specific adsorption sites but does not have H_2O/CO_2-sharing sites will be introduced. It readily adsorbs both H_2O and CO_2 from the humid flue gases and atmosphere, but the adsorbing H_2O does not interfere with the adsorption of CO_2. It is also highly stable after adsorption of H_2O and CO_2 because it was synthesized hydrothermally. (*Science* **2015**, *350*, 302-306)

The effective removal of $^{137}Cs^+$ ions from contaminated groundwater and seawater and from radioactive nuclear waste solutions is crucial for public health and for the continuous operation of nuclear power plants. Various $^{137}Cs^+$ removers have been developed, but more effective $^{137}Cs^+$ removers are still needed. A novel microporous vanadosilicate with mixed-valence vanadium (V^{4+} and V^{5+}) ions, which shows an excellent ability for Cs^+ capture and immobilization from groundwater, seawater, and nuclear waste solutions will be introduced. This material is superior to other known materials in terms of selectivity, capacity, and kinetics, and at very low Cs^+ concentrations, it was found to be the most effective material for the removal of radioactive Cs^+ ions under the test conditions. This novel vanadosilicate also contains hexadeca-coordinated Cs^+ ions, which corresponds to the highest coordination number ever described. (*Angew. Chem. Int. Ed.* **2014**, *53*, 7203–7208)

During the reprocessing of spent nuclear fuel rods, a highly moist off-gas mixture containing various volatile radioactive species such as iodine (I_2), organic iodides and nitric acid, is produced. Efforts have been made to devise materials, which can effectively capture radioactive iodine (I_2) and organic iodides from the off-gas without being damaged by moisture, nitric acid, and I_2. We observed that the hydrophobic all-silica zeolite, silicalite-1, is stable in 5 M nitric acid and adsorbs I_2, CH_3I, and CH_3CH_2I from highly acidic off-gas mixtures to a much greater extent than does activated carbon. Moreover, we found that I_2 forms a unique semiconducting three-dimensional supramolecular network within the silicalite-1 channels. The conductivity of the fully I_2 loaded silicalite-1 is observed to be ca. 10^4 S m^{-1}, which is ca. 10^8-fold higher than that of solid I_2. (*Energy Environ. Sci*. **2015**, *9*, 1050-1062)

In addition to the above a novel vanadosilicate which shows a high specificity to $^{90}Sr^{2+}$ capture and a microporous titanosilicate material for CO_2 capture from the air will be introduced.