H_2/D_2 separation at room temperature using solid-state dihydrogen complexes

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Hydron isotopes such as deuterium (D) and tritium (T) play important roles not only in the fundamental research but also in the industry. D has been used as NMR solvents, tracer, moderator in nuclear reactors, and silicon semiconductive devices. D and T are also used as the nuclear fusion reactors, which will be expected as a future energy supply. T is contaminated with water in the nuclear waste, and its removal will be beneficial from the viewpoint of environmental protection.

Natural abondance of D is 0.015 % (mainly in the sea water) and distillation process at 20 K is used to concentrate D to produce D_2 gas, which is energy-intensive process. Recently, quantum sieving has been extensively studied as a new technique of the hydrogen isotope separation. Beenakker and co-workers proposed the concept of kinetic quantum sieving (KQS), which utilize the difference of the diffusion rate in the small pore due to the thermal de Broglie wavelength between H and D. More recently, chemical affinity quantum sieving (CAQS) has been studied, which utilizes the difference of zero-point energy (ZPE) and resultant adsorption enthalpy (ΔH) among the hydrogen isotopes. Although CAQS potentially enables the hydrogen isotope separation at higher temperatures, the operating temperature was still below 200 K due to the small ΔH.

From the viewpoint of ΔH, metal-dihydrogen complex is one of the most appealing candidates for this target. The first metal-dihydrogen complex was reported by Kubas and coworkers in 1984, and hundreds of compounds were reported so far. In the metal-dihydrogen complexes, chemical bond is formed between metal and dihydrogen molecule, whereas H-H bond is kept albeit it is elongated and weakened. This can be regarded as an intermediate of physisorption and chemisorption, and therefore, moderate adsorption enthalpy and fast ad/desorption kinetics could be achieved. In spite of such advantages, there has rarely been studied on the hydrogen adsorption of the dihydrogen complexes in the solid state.

We have recently studied the hydrogen adsorption of solid-state dihydrogen complexes and found that ΔH of D_2 is significantly larger than that of H_2 in the most of dihydrogen complexes. In the presentation, we discuss the possible mechanisms of this phenomenon. In addition, we also report the demonstration of the separation of H_2 and D_2 using column chromatography at ambient temperature.