A Hard X-Ray Study of a Manganese-Terpyridine Dimer Catalyst in a Chromium-Based Metal Organic Framework

Alex Ramsey

Much of today's scientific research is geared toward discovering clean energy from renewable sources. A promising approach is to split water into its fundamental elements – hydrogen and oxygen. The hydrogen produced from this process is a source of clean energy. Plants carry out this water splitting routinely as a part of photosynthesis, but we cannot do this readily as the reaction requires a lot of energy, negating the value of cleaner and plentiful energy from hydrogen. Adding a catalyst provides an alternative pathway for the reaction so that it can proceed with lower energy input. Many such catalysts have been developed; the one used in this study is manganese based and is called manganese-terpyridine dimer (MnTD). The problem is that when individual molecules of MnTD interact they destroy each other, which limits practical use of the catalyst. However, by placing the catalyst in a Metal Organic Framework (MOF), the molecules of MnTD can be separated so that, in principle, they do not interact with each other and only interact with water. The MOF is essentially a repetition of boxes, and each box should contain one MnTD molecule. However, preliminary studies showed that a different molecule, not MnTD, may be inside the boxes. My research focused on identifying which molecules are present in the MOF. The primary method of analysis used was x-ray powder diffraction, a technique in which x-rays are scattered by a sample, and the intensity of the scattered radiation is measured as a function of scattering angle. Three samples were analyzed; one contained only the MOF, one contained the MOF and catalyst before catalysis, and one contained the MOF and catalyst during catalysis. The diffraction experiments were carried out at the Stanford Synchrotron Radiation Lightsource (SSRL) at SLAC National Accelerator Laboratory. Initial results revealed that the MOF structure falls apart during catalysis. To further analyze the data, I used several computer programs, including GSAS II, FOX, and VESTA. These programs allowed me to create maps that showed regions of electron density within the MOF and gave me clues as to which molecules were present. I found that MnTD was not the catalytic molecule in the MOF. The preliminary studies showed that manganese oxide is likely present, which is consistent with by data. However, the catalytic

rate for this system is much higher than that reported for manganese oxide, and thus future studies are needed to verify the identity of the catalytic molecule and determine the reasons for its unusually high rate of catalysis.