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Permalink
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Publication Date
2017-03-01

DOI
10.1016/j.gca.2017.01.019

Peer reviewed
Acceptance of the 2016 C.C. Patterson Award by William H. Casey

Madam President, Colleagues, Ladies and Gentlemen:

This award is precious to me because I lecture each year about the life and accomplishments of Claire Patterson to my freshman students. He was the most important geochemist of the 20th century and remains a model for us all of passion in research for the greater good of society (McGrayne, 2002).

We've seen aqueous geochemistry evolve to a field that emphasizes molecular properties and reaction kinetics from one of thermodynamics and box models. The key step was developing experimental models that are at the same scale as our simulations, so that we are not trapped in the futility of running experiments on bulk materials and then interpreting the results using molecular-scale simulations. Molecular understanding requires molecular models for both experiment and simulation and NMR spectroscopy probes reactions at the convenient time scale of microseconds to hours.

If I have any skill as a scientist, it is the ability to identify creative people with whom to collaborate and this address gives me a chance to showcase their accomplishments. As mentioned in the citation, Prof. Tom Swaddle showed us how to study ligand-exchange reactions, including via high-pressure NMR, along with Prof. Leone Spiccia of Monash University. When I say 'we', I mean myself and Brian Phillips, who is my very good friend but also the person who first noticed that the $^{17}$O-NMR peak assignments were wrong in the $[\text{AlO}_{4}\text{Al}_{12}(\text{OH})_{24}(\text{OH}_{2})_{12}]^{7+}$ molecule, allowing us to make measurements of oxygen-isotope-exchange rates in these clusters (Phillips et al., 2000) and expand them to other key classes (Casey and Rustad, 2007, 2015, Rustad and Casey, 2012). The suggestion that this molecule could serve as a molecular model for reactions at mineral surfaces was made by Geri Furrer, Bernhard Wehrli and Eric Wieland in an inspired paper (Wehrli et al., 1990). Brian and I found that single-atom substitutions, well away from the reaction, exert an enormous influence over the kinetics (Casey et al., 2000, Casey and Phillips, 2001, Lee et al., 2002).

Jim Rustad explained the experimental observation and identified a set of similar reaction steps and concerted motions of many atoms that were involved in the dissociation and oxygen-isotope exchanges in condensed nanometer-sized oxide ions. These steps explain the amphoteric distribution of rates, the averaging effect of small changes in composition, and the fact that regiospecific site exchanges, and complete dissociation of the molecules, have a similar dependence on solution composition. He showed that formation of a metastable intermediate is key and that this reaction proceeds via a predictable set of steps (Rustad and Casey, 2012). This 'Rustad Mechanism' has proven to be very robust (Casey and Rustad, 2007, 2015, Rustad and Casey, 2012).

This leads me to the Potato Cannon high-pressure NMR probe that allows geochemists to conduct NMR experiments on solutions to 2.0 GPa, and probably to 3.0 GPa (Pautler et al., 2014, Ochoa et al., 2015). It is so easy to use that undergraduates can make meaningful measurements. Help for this design came from colleagues in my Chemistry and Physics Departments at UCD, and in our NMR center.

Finally, I wish to point out that, although this is my first trip to Yokohama, it has always loomed large in my imagination. My mother was an Army nurse in 1951 stationed in Yokohama, where she met her future husband, a soldier in the Korean War. They had seven children, and her wedding kimono, made for her by a Japanese friend in Yokohama, hangs on my living-room wall today.

I thank our hosts in Yokohama.
References


