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Catalytic hydrogenation of non-polar unsaturated organic molecules promoted by bifunctional iron complexes

The hydrogenation reaction of unsaturated bonds promoted by a transition metal catalyst is one of the most fundamental transformations in organic synthesis. Using a metal-ligand bifunctional catalyst of transition metals is one of the attractive approaches. Shvo complex is the first example of a metal-ligand bifunctional catalyst that has Ru-H and O-H functional groups. The complex showed effective hydrogenation activity for organic molecules with a polar unsaturated bond such as ketone or aldehyde, but did not show activity for those with a non-polar unsaturated bond such as alkene or alkyne. This report shows preparation and isolation of new hydride iron complexes bearing a Si-H or a Ge-H bond tethered to the Cp derivative ligand and their hydrogen transfer activity towards unsaturated organic compounds. Iron hydride complexes bearing a Si-H or a Ge-H functional group in a ligand, [2,5-SiPh$_3$-3,4-butylene-(ή^5-C$_4$COEEt$_2$H)]Fe(CO)$_2$H (E=Si: 1, Ge: 2) and [2,5-SiPh$_3$-3,4-butylene-(ή^5-C$_4$COSiMe$_2$OSiMe$_2$H)]-Fe(CO)$_2$H(3), were synthesized in the reaction of iron acetonitrile complex, [2,5-SiPh$_3$-3,4-butylene-(ή^5 -C$_4$CO)]Fe(CO)$_2$(NCCH3), with Et$_2$EH$_2$ or HSiMe$_2$OSiMe$_2$H. Conventional bifunctional complexes which possess O-H on the ligand and M-H bonds have been reported to react predominantly with ketone or aldehyde rather than with alkyne or alkene. In stark contrast, 1 did not react with methyl phenyl ketone, but 1 reacted with p-tolylacetylene to give the corresponding alkene and alkane (Scheme 1). The reactivity of 1 is considered to be attributable to the polarity of the Si-H bond being opposite to that of an O-H or N-H bond. This is the first example of iron promoting transfer hydrogenation reaction from alcohol to alkyne or alkene in one pot. Such catalytic transfer hydrogenation reactions have been reported for Ru, Rh, Ir, and Pd, and most of them require addition of base such as K$_2$CO$_3$, Na$_2$CO$_3$, and KOH.

Biography

Hiroshi Nakazawa has completed his PhD in 1981 from Hiroshima University, Japan. After working at Tokyo Institute of Technology and at University of Utah as a Post-doctoral Research Fellow, he became a Research Associate at Hiroshima University in 1984, and was promoted as Associate Professor. From 1994 to 1996, he was appointed an Associate Professor of Institute for Molecular Science. Since 2002, he has been a Full Professor of Osaka City University. He received the Commendation for Science and Technology from the Ministry of Education of Japan in 2009, the Award for Chemical Education from the Chemical Society of Japan in 2015, and Japan Society of Coordination Chemistry Award in 2016.

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