Formation, stability, and reactivity studies of neutral iron sulfide clusters

SHI YIN, ZHECHEN WANG, ELLIOT BERNSTEIN, Chemistry Department, Colorado State University — Different methods are used to generate neutral iron sulfide clusters to study their formation, stability, and reactivity, employing a time of flight mass spectrometer (TOFMS) with VUV (118 nm) radiation single photon ionization (SPI). Neutral Fe$_{m}$S$_{n}$ ($m = 1-4, n = 1-6$), and hydrogen containing Fe$_{m}$S$_{n}$H$_{x}$ ($x > 0, n > m$) clusters are generated by the reaction of seeded H$_2$S in a helium carrier gas with laser ablated iron metal within a supersonic nozzle. The observed strong signal of association products Fe$_2$S$_2$(SH)$_{0,1}M$ ($M = \text{CO, C}_2\text{H}_4, \text{C}_3\text{H}_6$) suggest that the Fe$_2$S$_2$(SH)$_{0,1}$ clusters have the high activity for interactions with these small molecules. In order to avoid the effect for reactivity from hydrogen containing clusters, pure Fe$_{m}$S$_{n}$ clusters are generated through laser ablation of a mixed iron/sulfur target in the presence of a pure helium carrier gas. (FeS)$_m$ ($m = 1-4$) is observed to be the most stable series. Reaction of CO and H$_2$ on neutral (FeS)$_{1,2}$ clusters is farther investigated both experimentally and theoretically. A size dependent reactivity of iron sulfide clusters toward CO is characterized. The reaction FeS + CO → Fe + OCS is found for the FeS cluster. Products Fe$_2$S$_2^{13}$COH$_2$ and Fe$_2$S$_2^{13}$COH$_4$ are identified for reactions of $^{13}$CO and H$_2$ on Fe$_2$S$_2$ clusters: this suggests that the Fe$_2$S$_2$ cluster has a high catalytic activity for hydrogenation reactions of CO to form formaldehyde and methanol. DFT calculations are performed to explore the potential energy surfaces for the two reactions: Fe$_2$S$_2$ + CO + 2H$_2$ → Fe$_2$S$_2$ + CH$_3$OH; and Fe$_2$S$_2$ + CO + H$_2$ → Fe$_2$S$_2$ + CH$_2$O.

Elliot Bernstein
Chemistry Department, Colorado State University

Date submitted: 13 Nov 2013
Electronic form version 1.4