NANO HIGHLIGHT
A New Class of Oxidation Catalysts: The Role of Atomically Dispersed Metals in Nanostructured Oxides

NSF NIRT Grant 0304515

PI: Maria Flytzani-Stephanopoulos(1)
Co-PI: Terry E. Haas
Co-PI: Regina Valluzzi
Co-PI: Howard Saltsburg
Department of Chemical & Biological Engineering
Tufts University Medford MA 02155

The need for high purity hydrogen for use in low temperature fuel cells has led to a resurgence of interest in purification of hydrogen derived from processing carbon based feedstocks. A common process used to reduce the carbon monoxide concentration of the process gas to an acceptable level is the water gas shift reaction (WGSR) . In the reaction, carbon monoxide can be combined with water to form carbon dioxide and hydrogen over an appropriate catalyst. Many studies over the last ten years showed that gold nanoparticles supported on various oxides are effective catalysts for the WGSR. In order to design and optimize such a catalyst it is important to know the details of the chemical processes involved in the reaction. A number of proposals have been offered to explain the process but all depend on the properties of the metallic nanoparticle.

Recent work in this laboratory has demonstrated that for the system of nanoparticles of gold on nanocrystalline cerium oxide, **the gold nanoparticles do not participate directly in the WGS reaction but are merely spectators** (2). The active catalytic entity is nonmetallic: either a cluster of a few atoms or more likely gold ions. These entities are formed on nanocrystalline cerium oxide, during the initial preparation but once the particles are removed there is no effect on the process. The removal of 90% of the gold that was initially present in the form of metallic particles suggests that the economics of the catalytic process can be improved substantially. This finding is the basis for the work to be carried out under the NIRT grant funded effective 8/15/2003.

We will be examining other nano-metal particle based catalysts and other reactions to determine whether or not the findings for gold on ceria represent a more general phenomenon. If so, there are significant economic benefits when any precious metal catalysts are to be used. To examine the role of the oxide in the reaction, in addition to studying several other nanocrystalline oxides, we will utilize the results of a recent finding at Tufts that showed that it is possible to place an isolated ion on a carbon based structure (3). Studies of such an entity without the oxide could clarify the role of the oxide. Although several possible structures for the catalytic entity can be described it is important to determine the relative stability of the proposed structures. To that end we will be carrying out relevant quantum mechanical calculations. Finally, the role of the interface between the nanometal particle and the support is unclear. Fabricating controlled structures with well defined interfaces may shed light on the mechanism of formation of the catalytic entity (4).

References

[1] For further information about this project: email **<mflytzan@tufts.edu)>**
[2] Qi Fu, Howard Saltsburg, Maria Flytzani-Stephanopoulos, *Science* 301 935 (2003)
[3] Regina Valluzzi, R. Guertin, Terry Haas MRS (S29.011) March 2003
[4] Ioannis Zuburtikudis, Howard Saltsburg, *Science* 258 1337 (1992)