Biomass Catalyst Capabilities

Catalyst Synthesis Facilities

Atomic Layer Deposition (ALD) Synthesis Facilities

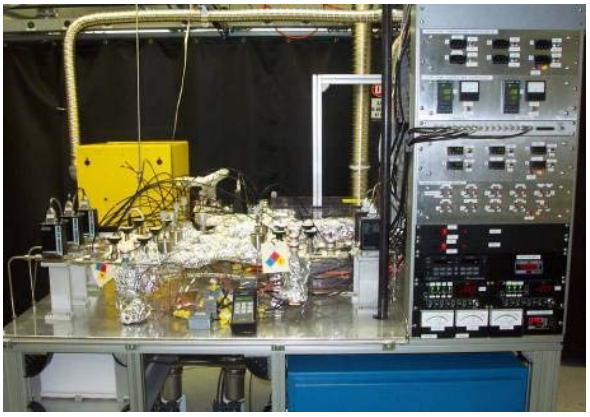
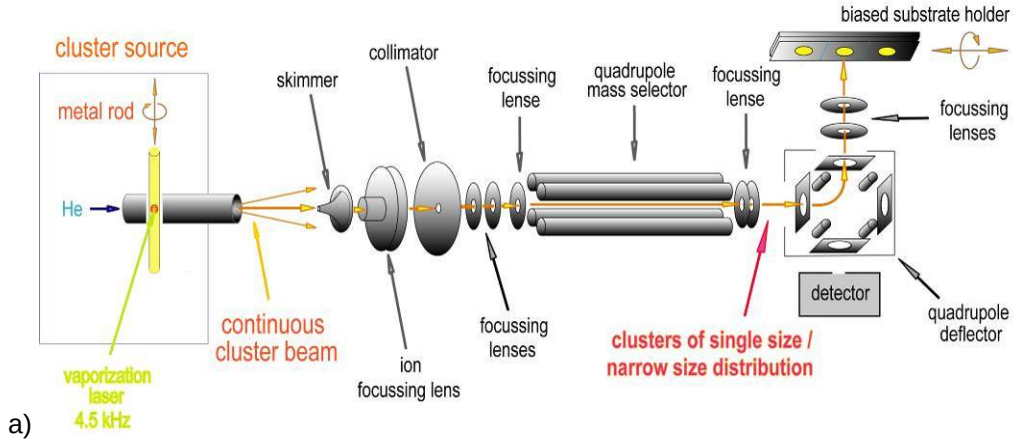


Figure 1. Viscous flow ALD reactor at Argonne

We currently have three ALD viscous flow reactors operating at Argonne in two separate laboratories. These unique instruments were designed and constructed at Argonne, and each has ten independent precursor delivery channels allowing a wide variety of materials to be deposited including complex oxides, superlattice layers, metals, and metal alloys. These flexible systems allow us to deposit a wide range of materials with atomic level precision onto nearly any substrate including high surface area powders and single crystals. The Argonne ALD reactors have in situ quartz crystal microbalance and quadrupole mass spectrometer measurement capabilities enabling detailed studies of new ALD chemistries. Our ALD laboratories are also equipped with a range of thin film analytical instruments including variable angle spectroscopic ellipsometry, x-ray photoelectron spectroscopy, x-ray fluorescence spectroscopy, Fourier transform infrared analysis, four-point probe resistivity measurements, stylus profiling, Raman microanalysis, and ultraviolet-visible spectrophotometry. We also have ready access to x-ray diffraction analysis, atomic force microscopy, and scanning electron microscopy (with backscattered electron detection and EDAX).

Size-selected cluster facility. The continuous beam of metal clusters is generated in a new high-flux laser vaporization cluster source which utilizes a Quantronix Nd:YAG laser operating at 4.5 kHz (see Figure 3). The beam of neutral and charged clusters passes through a biased skimmer into the ion guide of the second differentially pumped vacuum stage and then into the third vacuum stage. The positively (or negatively) charged clusters are guided and focused into the quadrupole mass spectrometer (Extrel) for mass analysis. Narrow cluster distributions in the mass range of 1-2000 amu with a few dominant sizes can be produced by optimizing the temperature of the clusters source, pressure of the helium carrier gas and potential settings on the individual ion optics elements. For example, in the case of gold and platinum clusters, Au_n and Pt_n clusters in the size range n=1,…10, with one to four dominant sizes were produced (see references in text). After the desired cluster distribution is produced, by reversing the polarity settings on the quadrupole deflector, the cluster ions are deflected into an ion lense setup placed in front of the substrate mounted on a translation stage. The kinetic energy of clusters landing on the surface is controlled by biasing the substrate. Typical deposition energies are in the order of 0.5 eV/atom. The flux of clusters landing on the support is monitored in real-time using a picoammeter (Keithley). Typical deposition currents are in the order of tens to hundreds of picoamperes, depending on the size of clusters. Maximal cluster currents in the order of up to several nanoamperes are achievable. Currently, an upgrade of the apparatus is under progress with the goal to expand the size/mass range.



a)

b)

c)

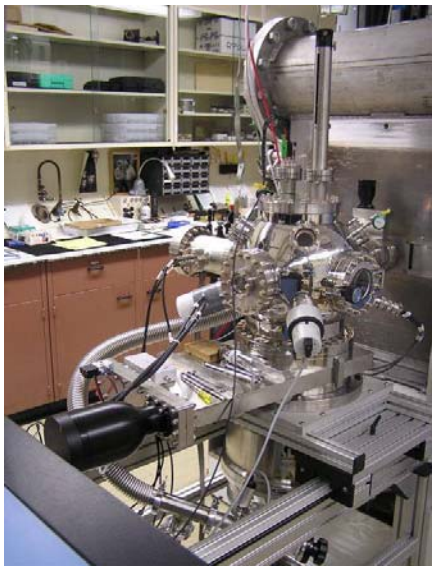


Figure 2. (a) Schematic of the size-selected cluster deposition setup, (b) electronic control unit of the mobile deposition stage and (c) closeout of the deposition chamber

In addition to pure metal clusters, bimetallic or composite clusters can be produced by using an alloy target rod and seeding the carrier gas with reactive compounds, respectively. The recently acquired software allows for a selection of clusters of defined size and composition with atomic (isotopic) precision, a key element for the proposed studies on bimetallic model nanocatalysts.

In the case of flat surfaces precoated with thin ALD oxide films, surface coverage of 2-5 % ML will be employed, with the goal to avoid aggregation of clusters into larger particles after they land on the surface. Deposition of size-selected clusters has been recently performed on with ALD precoated AAO membranes and the catalytic activity of the $Pt_{8\text{-}10}$ and $Co_{25\text{-}32}$ cluster-based catalysts successfully tested in oxidative dehydrogenation of propane and decomposition of a cellulose surrogate, respectively .

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