Volatilization Behavior of Supported Au Nanoparticle Arrays Under H_2 at High Temperature

Supporting Information

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XPS Measurements

XPS spectra were acquired following each treatment to measure the Au 4f /Si 2p peak area intensity ratios reported in the main body of the manuscript. Survey spectra and Si 2p spectra were nearly identical for all samples. Representative spectra are plotted below. There was a noticeable change in Au 4f peak intensity as the treatments progressed, especially with the initial treatment in O_2 and the first treatment in 10% H_2, and then again at 800 °C. A plot of the Au 4f spectra normalized to the intensity of Si 2p illustrate this change.

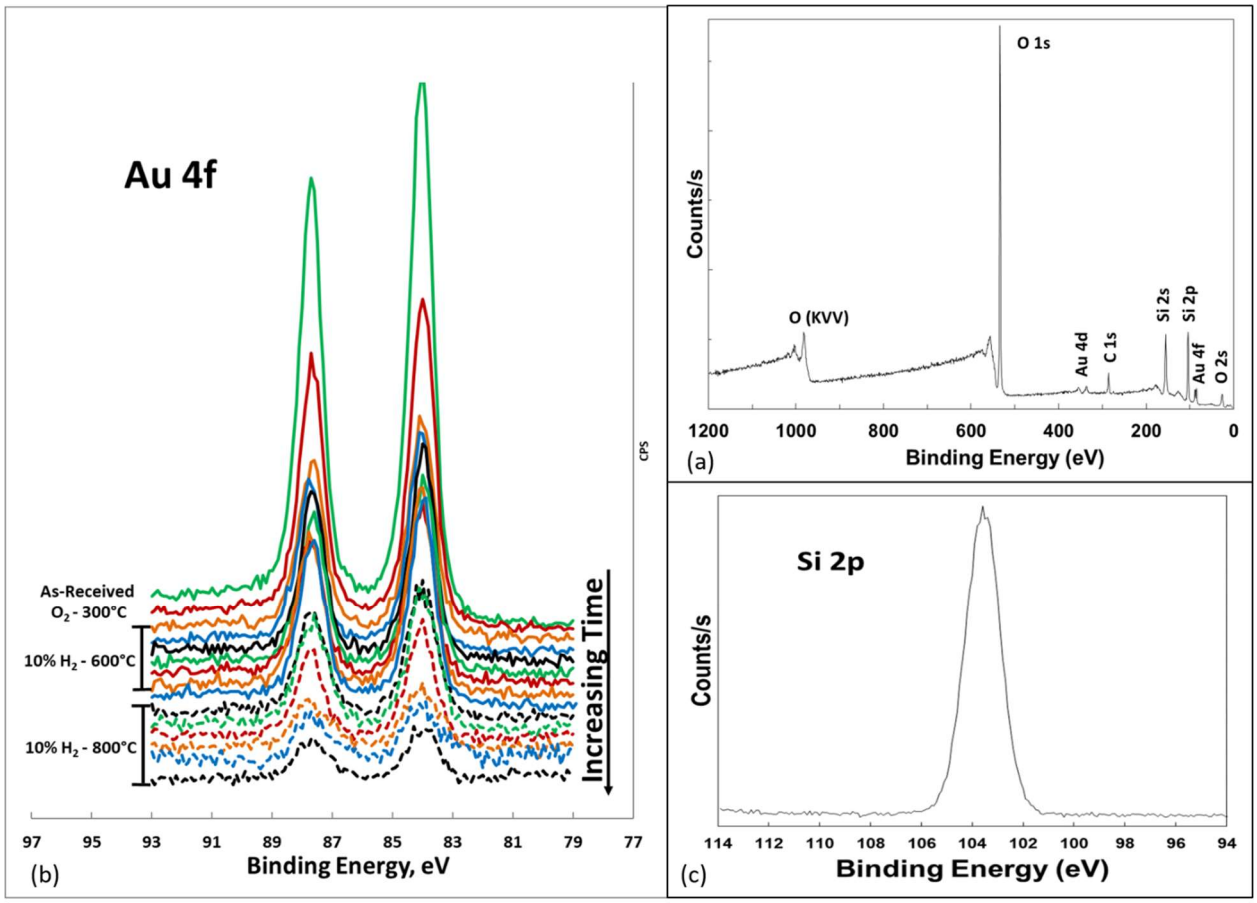

Figure S1. XPS spectra for Au/SiO_2. (a) representative survey scan, (b) Au 4f spectra normalized to Si 2p intensity as a function of treatment time under various gas exposures; 800 °C treatments are represented with dashed lines, (c) representative Si 2p spectrum.

Model Uncertainties and Error Analysis

Table 1 lists the parameters used for the exposures in 10% H_2/N_2. Because of the complexity of calculations, a strict analysis of error is beyond the scope of this work. The largest uncertainties are in the D_{AB} term, which was calculated using Eqs. 8 and 9. The other parameters are much more accurate in comparison, being derived from literature values (as referenced) using less complex calculations. The P_{Au} and P_{AuH} terms can vary by orders of magnitude with T, while the other terms much less so. For example, γ values are ± 0.06 J/m^2. The overall effects on the

model curves in Fig. 1 are that the 600 °C curve will remain pretty flat regardless and should have small uncertainties while the 800 °C part of the curve would have larger error bars since the slope of the curve is proportional to flux, which is proportional to D_{AB}. The error bars for the 800 °C part of the curve would get larger with each step as each time increment would propagate the errors further.

For the experimental data, the precision of the SEM values is very good with a average relative standard deviation, based on the standard deviation for 20 measurements for each point (time/temperature treatment), of ± 1.2 nm with the largest standard deviation of ± 2.0 nm being found for the measurements after the final 800 °C treatment. This would make the error bars in Fig. 1 about the same size as the circle used to plot the data. For XPS Au 4f/Si 2p intensity ratios, the standard deviation for 3 measurements taken after the final treatment at 800 °C, where the error would be expected to be the greatest, was ± 11%.

Au Evaporation Model at 700°C

The Au evaporation model was used to predict how the size of Au nanoparticles would behave if the temperature were switched to 700 °C instead of 800 °C after 80-hour exposure to 10% H_2/Ar at 600 °C. The rate of Au loss is predicted to be comparatively much slower at 700 °C instead of 800 °C.

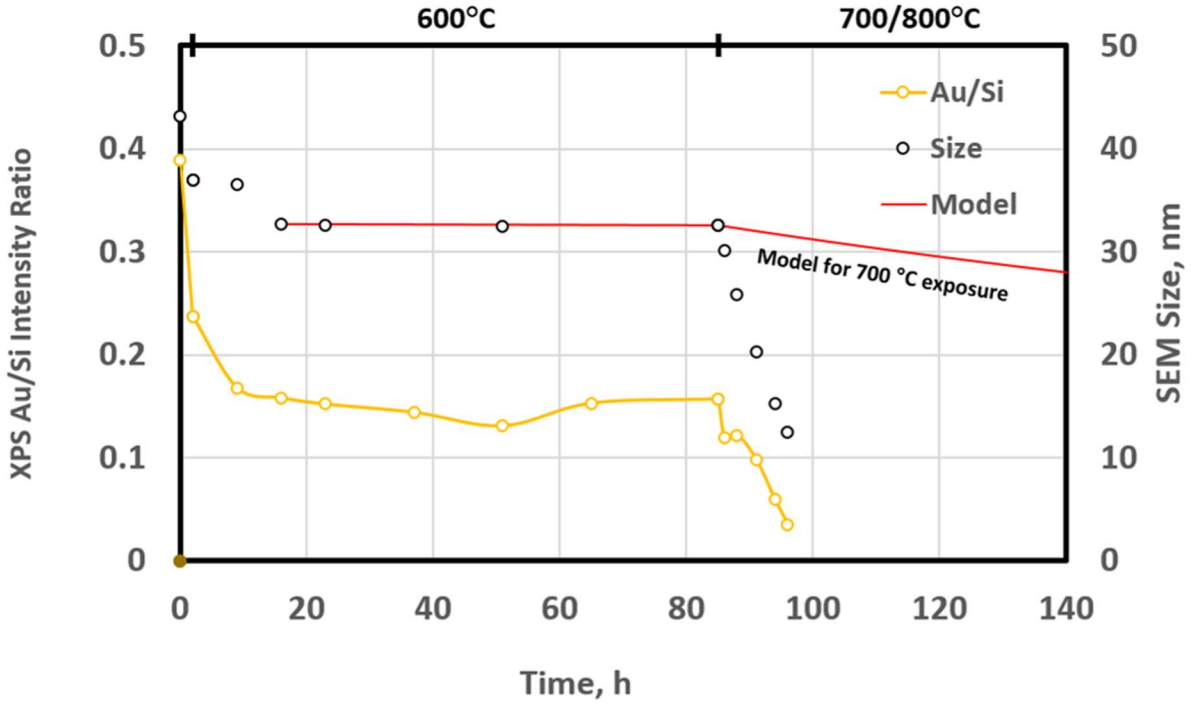

Figure S2. Model-predicted Au nanoparticle size for 600 °C initial exposure followed by 700 °C exposure after 85 hr total treatment time versus measured XPS Au/Si intensity ratio and SEM nanoparticle diameter during exposure of the Au nanodot in N_2-10% H_2 at 600 °C (2-85 h) and at 800 °C after that. The initial exposure was in O_2 at 300 °C (0-2 h).