Supporting Information

Real-Time Monitoring of the Aging of Single Plasmonic Copper Nanoparticles

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Experimental Section

Single NP Dark-field Imaging and Scattering Spectroscopy Measurements

The dark-field measurements were carried out on an inverted microscope (eclipse Ti-U, Nikon, Japan) equipped with a dark-field condenser (0.8 < NA < 0.95), a true-color digital camera (Nikon DS-fi), and a 300 mm focal-length and 300 grooves/mm monochromator (Acton SP2300i) with a 512×512-pixel cooled spectrograph CCD camera (CASCADE 512B, Roper Scientific). A few-micron-wide aperture was placed in front of the entrance slit of the monochromator to keep only a single NP in the region of interest at the grating dispersion direction. The true-color scattering images of NPs were taken using a 40X objective lens (NA = 0.8) and the true-color camera with a white light illumination from a 100 W halogen lamp. The image acquisition software is NIS-Elements provided by Roper Scientific. The scattering spectra of NPs were taken using the same optics, but they were routed to the monochromator and spectrograph CCD. The scattering spectra from the single NPs were corrected by subtracting the background spectra taken from the adjacent regions without the NPs and dividing with the calibrated response curve of the entire optical system. The integration time used in all experiments spectral acquisitions is 10 second.

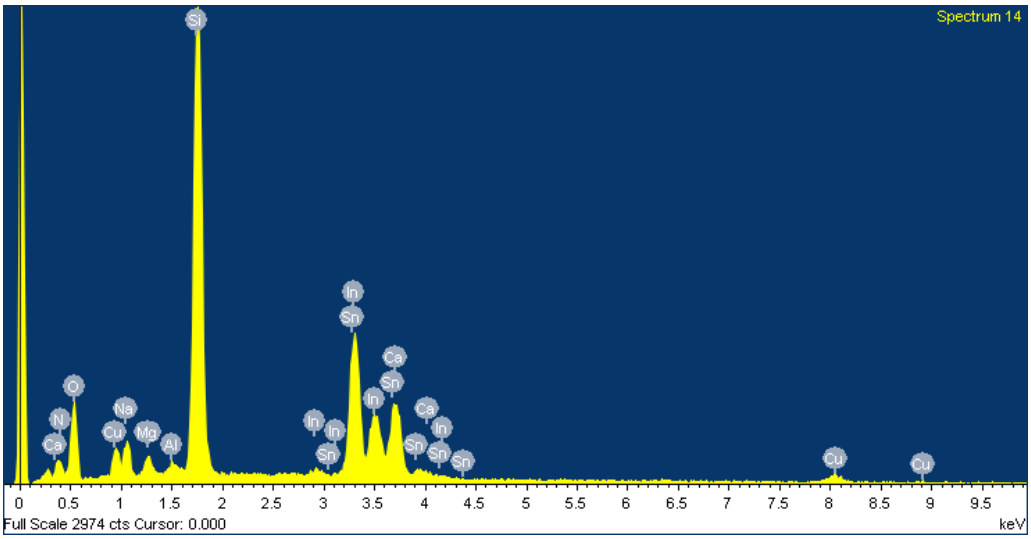
Cleaning Procedure of Microscopy Slides

Cleaning of microscopy slides surfaces was accomplished by rinsing the slides with methanol, allowing them to dry, and then placing them in a UV ozone cleaner (Jelight Co. model 342) for 25 min on each side. Slides were checked using water

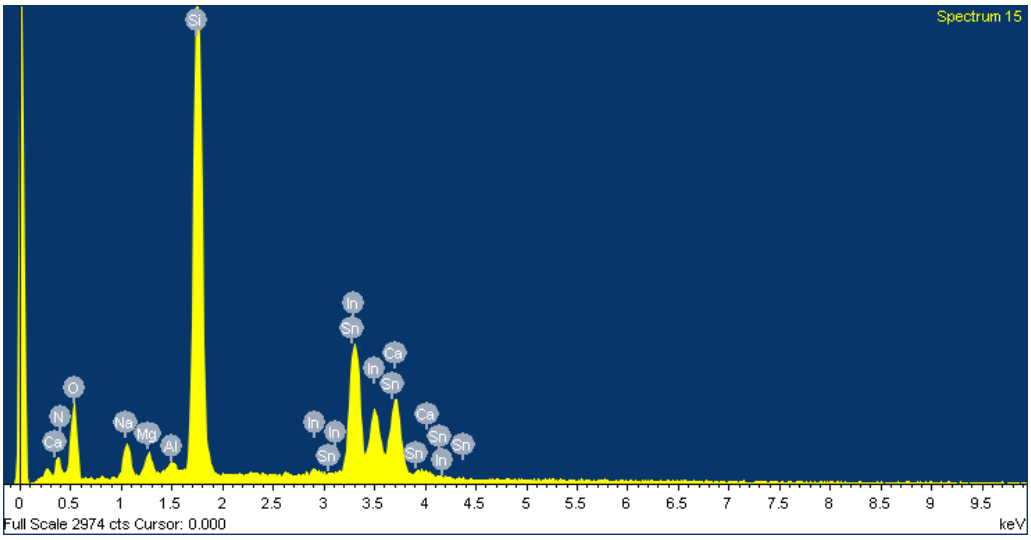
contact-angle measurements to determine cleanliness; a contact angle of 5° indicated that slides were sufficiently clean to produce uniform silane monolayers with minimal fluorescence background.

Characterization

Energy Dispersive Analysis of X-Ray (EDAX) were imaged by an FEI-Sirion 200 field emission (FEI Co. with 20 kV operating voltage). Spectra from the EDAX show the elemental composition of a material in a qualitative way. The material is placed in a scanning electron microscope and bombarded with a beam of electrons, typically about 20kV.



ESI 1. EDAX spectrum of Cu oxide layers on ITO substrate.



ESI 2. EDAX spectrum of bare ITO substrate.

Table 1. The content analysis of the electrodeposition layer on ITO and bare ITO substrate

Element	Atomic% (copper oxide layer on ITO)	Atomic% (bare ITO)
O K	50.98	48.79
Cu K	1.09	0
Na K	3.77	4.11
Mg K	1.37	2.01
Al K	0.43	0.99
Si K	25.65	25.81
Ca K	2.97	3.54
N K	6.48	7.12
In L	6.64	6.76
Sn L	0.62	0.87
Totals	100.00	100.00