

Photo-CVD Coating of Nanoparticles with Silicon Dioxide

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Abstract

Extending previous research [1-6], the ability to coat nanoparticles with silicon dioxide (SiO_2) using a xenon (Xe_2) excimer lamp at standard atmospheric temperature and pressure was demonstrated using a photo-chemical vapor deposition (CVD) process. Sodium chloride (NaCl) was used as at the core particle, and tetraethyl orthosilicate (TEOS) in concentrations up to 0.70 sccm was provided as the SiO_2 precursor. NaCl particles of approximately 40-50 nm diameter received coatings of repeatable thickness after an exposure time of approximately 1 second in 172 nm light. The ability to control coating thickness by modifying TEOS flow rate was considered, and coating thickness was shown to increase with TEOS flow rate up to a maximum thickness of up to 2.0 nm in these conditions. Characterization showed the process provided a practically uniform covering of amorphous silica on approximately spherical particles.

Procedure

To verify the coating of particles, a straightforward tandem differential mobility analyzer setup (TDMA) was used. Sodium chloride, atomized with a nitrogen carrier gas, was dried and size selected using a differential mobility analyzer (DMA). Extra flow was vented to assure 1.0 slm through the DMA. The flow was mixed with varying quantities of tetraethyl orthosilicate (TEOS) with the appropriate amount of carrier nitrogen to maintain vapor pressure at the ambient 21°C.

The mixture then received 50 mW/cm² of 172 nm radiation from a Ushio UER20H-172C excimer lamp. Intensity was periodically verified using a Coherent Fieldmate laser power meter. A particle residence time in the illumination chamber of between 1.7-2.3 s was derived from an Ansys 10.0 flow simulation.

After receiving radiation, the aerosol passed through a charger, then a second DMA before entering a TSI 3025A condensation particle counter. Stepping through a range of voltages, the second DMA passed different mobility diameters to the counter, and provided a complete distribution of particles in the flow. Controls were created using the same setup with the lamp off, with TEOS varying between none and 0.70 sccm. To capture particles for characterization, electrostatic precipitation was used, with a voltage of 2.5 kV. For each collection, a standard Cu TEM grid was attached to the positive terminal of the precipitator. Additionally, for each collection used for the FTIR analysis, an uncharged stainless steel screen was placed directly across the flow path of the particles.

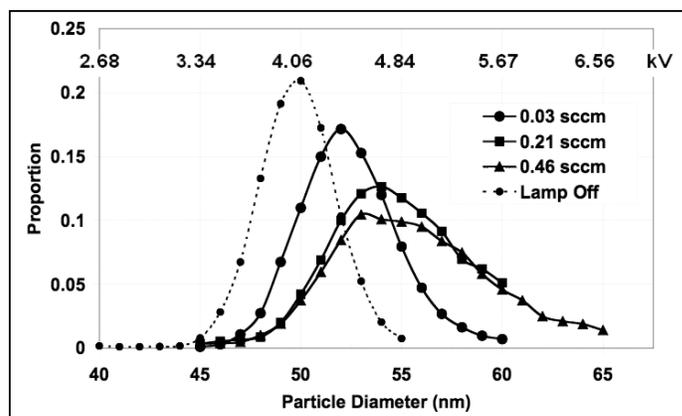


Figure 1: Mobility diameter by precursor quantity.

Results

The TDMA data demonstrate a size increase of the original particles as a result of the particle processing. Further TEM analysis indicates that the increased particle diameter is due to a coating process occurring as a result of photo-CVD. Both 40 and 50 nm particles experienced coating, however, most analysis was conducted on 50 nm particles because the atomizer produced a larger sample at that size.

In Figure 1, 50 nm particle results generated from the averages of five trials are presented. With increasing TEOS concentration, the particle coating thickness (half the difference between final and initial sizes) increased until a probable saturation thickness of about 2.0 nm. Thickness variation was approximately normally distributed, with more variance for larger TEOS flow rates. Tests without NaCl yielded minimal particles, implying that no self-nucleation of TEOS occurred.

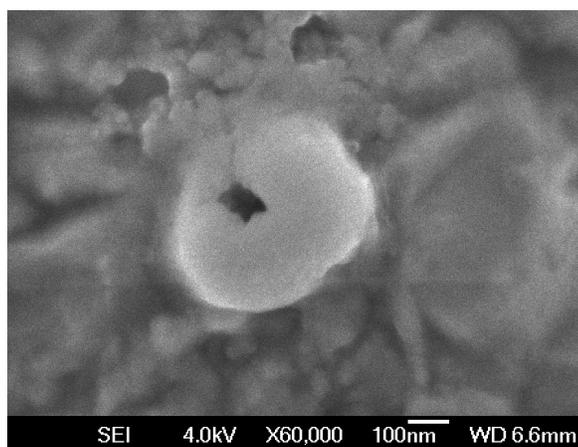


Figure 2: SEM image of an accumulation of coated nanoparticles.

Figure 2 displays a cluster of approximately spherical coated nanoparticles. There was evidence of some agglomeration, but individual particles of the appropriate size and consistent shape were clearly identifiable. This image was captured using a JEOL 6500 field transmission scanning electron microscope, operating with an excitation voltage of 4.0 kV, and a working distance of 6.6 mm. Figure 3, captured with a JEOL 1210 transmission electron microscope, clearly illustrates coatings of approximately uniform thickness despite a variety of vaguely spherical base particle shapes.

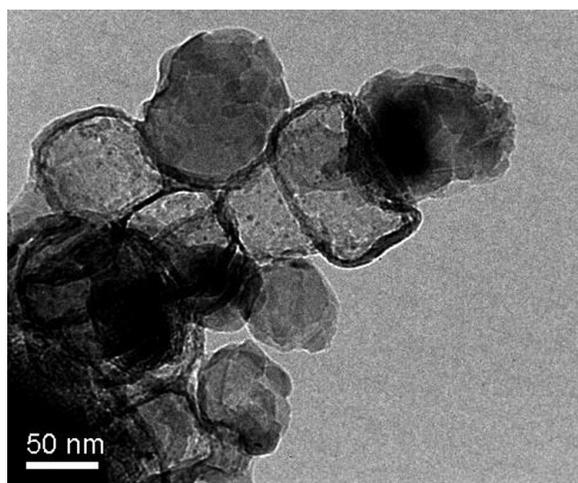


Figure 3: TEM image of nanoparticles and coatings.

X-ray diffraction characterization was conducted using a Bruker-AXS D5005, with a step size of 0.08 degrees 2- θ and 6.0 s step duration. Samples tested were polydisperse coated and uncoated particles, because no DMA was used. The results were analyzed using Jade 7.0, and no evidence of SiO₂ crystallization was found. Fourier-transform infrared radiation results were obtained with a Magma-IR 550 spectrometer.

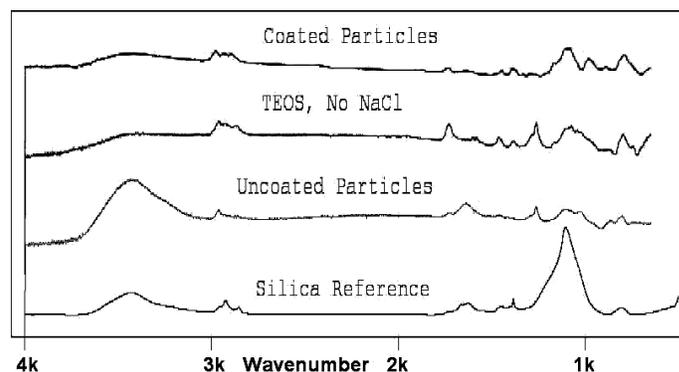


Figure 4: FTIR results.

Step size was 1 wave number with 30 samples per step, and the data was analyzed with Essential FTIR 1.20. Figure 4 shows that the difference between the coated particles and the uncoated control is only a few peaks, but each of those match a peak for silica, again providing strong evidence of coating. Particularly noticeable is the Si-O-Si asymmetric stretching peak at ~ 1120 wave number [5]. These silica peaks are more clearly pronounced in the coated particle data than the TEOS alone, demonstrating that the NaCl base contributes to silica formation.

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