

Effect of Synthesis Temperature on the Formation of GAC supported Pd and Au NPs

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Palladium (Pd) based catalysts are of increasing interest for the removal of volatile organic compounds (VOCs), such as trichloroethylene (TCE), from water by hydrogenolysis. Pd is often used in combination with promoter metals, such as gold (Au), which can increase TCE reduction rates by up to X-fold [1]. As a support, Granular activated carbon (GAC) has several advantages, including its capacity to absorb contaminants, which concentrates the reactants at the catalyst surface. It also provides a high surface area (porous) or anchoring Pd and Au nanoparticles (NPs). Previously, we have reported the synthesis of Pd/Au/GAC at 70°C to achieve the Pd and Au NPs on 20-40 mesh GAC catalyst, which has rapidly degraded TCE in laboratory batch experiments [2].

To determine how temperature during synthesis affects the Pd and Au NP morphology and distribution on the GAC, and the reactivity of the resulting material, the catalysts were synthesized at two temperatures, 70°C and room temperature (RT), using a 2:1 molar ratio of Pd to Au precursor, with solvent acetone. These temperatures were chosen since 70°C is slightly hotter than the boiling point of acetone (56°C), and RT (22-25°C) is the most convenient temperature condition to maintain. SEM equipped with EDS was used to characterize the surface of the GAC grains. Figure 1 (a) shows a survey of NPs uniformly distributed the surface of GAC synthesized at RT. (b) shows the that NPs appear to be of two sizes, the cluster of small NPs in the range of 10-20 nm and the larger particles appear to be in the range of 60-90 nm. (c) confirms the particles are composed of Pd and Au. Chlorine (Cl) and silicon (Si) are contaminants of GAC introduced during handling. Figure 2 (a) shows the surface of the GAC-based catalyst synthesized at 70°C, with (b) showing predominantly larger particles on the surface, in the range of 180–350 nm. Figure 2 (c) confirms these NPs are composed of Pd and Au. As observed previously [2], the larger particles appear to be Au-rich, while the smaller particles tend to be Pd-rich. Our observations suggest that some of the formations are bimetallic, while others are elemental NPs or heterogeneous structures, further identification of which is the target of future work. We hypothesize that the difference in sizes of these NPs, between the catalysts synthesized with two different temperatures, is due to an increase in aggregation of Au NPs at the higher temperature.

As-produced catalysts show no remarkable distinguishing features visible to the naked eye, as seen in Figure 3 (a). The effect of NPs' size on the activity of the catalyst was tested *via* degradation of TCE in a hydrogen-rich aqueous atmosphere. Samples were analyzed using a gas chromatographer with a flame-ionized detector (GC/FID), and are reported in Figure 3 (b). The RT samples degraded TCE faster than 70°C sample, possibly because the RT samples offer more of the catalytically active NP surface than their counter-part, despite the fact that the same amount of precursors were used for synthesizing both samples. Further investigations are ongoing towards better particle size control and aggregation, and its impact on degradation of contaminants. In general, NP aggregation control can have several benefits across different applications.

References:

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 [2] K Meduri, A Barnum, GO Johnson, PG Tratnyek, and J Jiao, *Microscopy and Microanalysis*, **22** (2016), p. 332

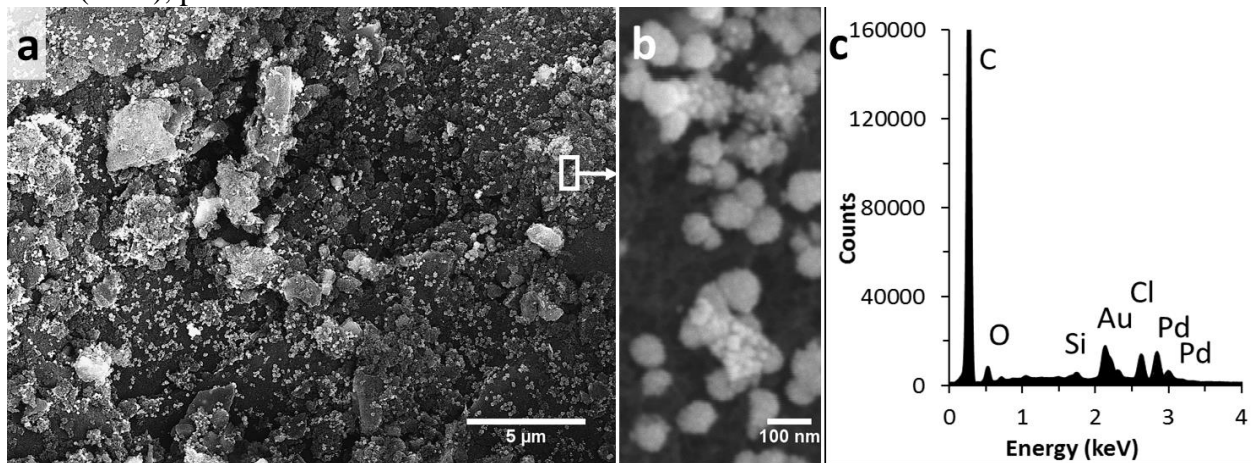


Figure 1. (a) SEM image at 11.5kX of the surface of the RT synthesized catalyst. (b) 220kX image showing the NPs morphology. (c) is the EDS spectrum confirming Pd and Au composition of NPs.

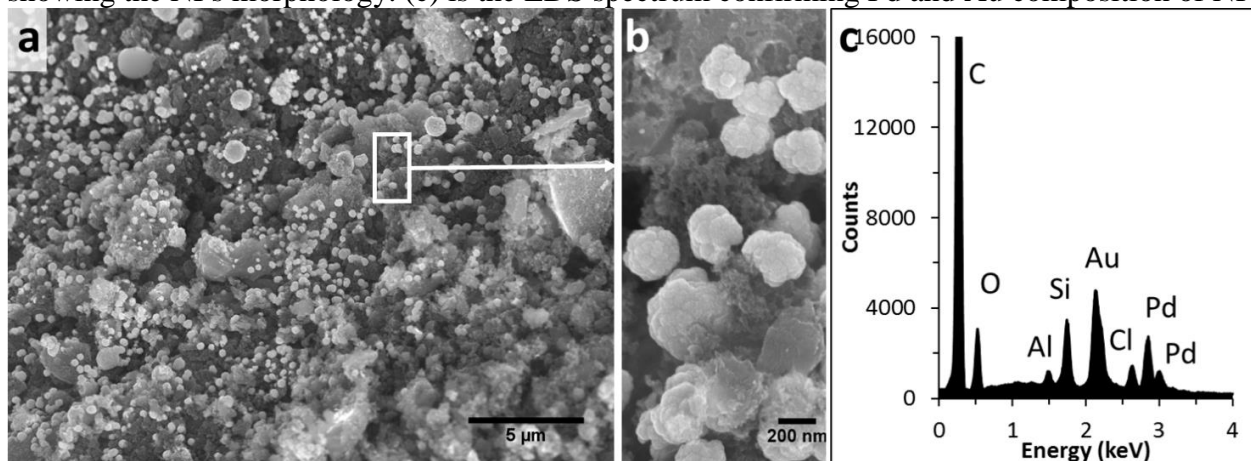


Figure 2. (a) SEM image at 11.5kX mag of the surface of 70°C synthesized catalyst. (b) 100kX image showing the NPs morphology. (c) is the EDS spectrum confirming Pd and Au composition of NPs.

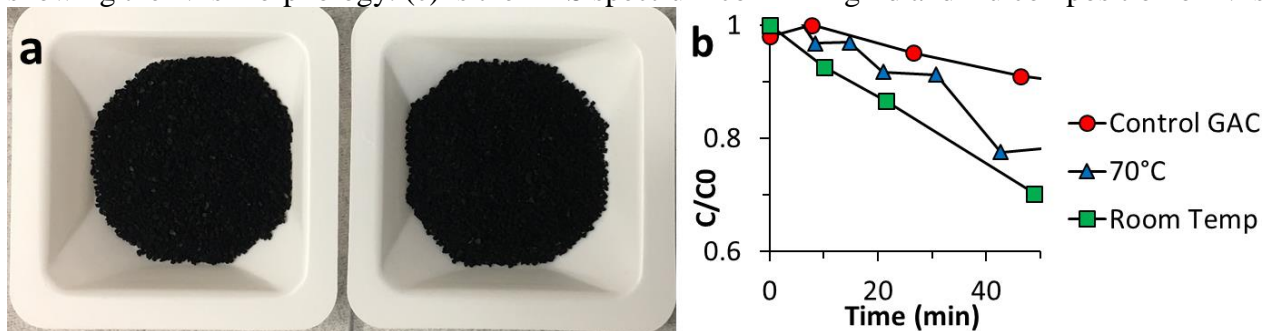


Figure 3. (a) Pictures of samples fabricated at room temperature (right) and 70°C (left). (b) shows the TCE degradation rates of the samples.