Characterization of Palladium and Gold Nanoparticles on Granular Activated Carbon as an Efficient Catalyst for Hydrodechlorination of Trichloroethylene

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Palladium (Pd)-based catalysts are of interest in a variety of water treatment applications. Studies have shown that hydrogenolysis catalyzed by Pd converts trichloroethylene (TCE) into ethane without the formation of vinyl chloride and other chlorinated intermediates. Gold (Au) has been observed to enhance the catalytic performance of Pd, and can change the reaction products formed [1]. Granular activated carbon (GAC) is often used as a filter for water purification due to its strong adsorption capability. Our novel catalyst combines the highly adsorbent nature of GAC with the catalytic Pd and Au nanoparticles (NPs) to form a bimetal catalyst for the decomposition of TCE. The resulting catalyst is formed by a two-step solvothermal process, using palladium (II) acetate and gold (III) chloride in the presence of a solvent to form nanoparticles on 20-40 mesh GAC. This catalyst has then been tested with aqueous TCE in batch experiments. After an equilibration period that appears to be dominated by mass transfer effects, the rate of TCE degradation is rapid and pseudo-first order. Figure 1 shows the batch test results in the presence of excess hydrogen at room temperature, compared to appropriate controls.

The Pd and Au NPs on GAC have been characterized here using SEM/EDS and HRTEM/STEM/EDS, while FIB was used for microscopy sample preparation. Pd and Au NPs were found to be distributed throughout the surface of GAC, as seen in SEM images in Figure 2, forming both individual NPs and congregating in other areas. Some pores of the GAC are several orders of magnitude larger in size than the NPs, as can be seen in the areas sectioned by the FIB in Figure 3, so it is possible that the NPs are deposited inside the GAC as well. Some of the nanoparticles were found remaining in the solvent after synthesis; this along with crushed catalyst were examined in the HRTEM, shown in Figure 4 (a) - (g). As seen in Figure 4 (a) and (b), crystals of Pd and Au were formed during synthesis, where Pd formed the smaller crystals in the range of about 1nm, but the Au tended to aggregate to bigger clumps of about 30 nm diameter. The EDS maps in Figure 4 (e) and (f) confirm the same information. On the GAC, the NPs can be seen as larger clusters in Figure 4 (g).

This type of catalyst has great potential to replace industrial catalysts and adsorbents as the more efficient alternative with a longer lifetime. Further optimization of the catalyst composition and structure is being carried out now. The influence of Au as a promoter for Pd catalytic performance is not well understood and is being examined systematically. Detailed investigations of the mechanism of decomposition and interaction between catalyst and contaminant TCE are also being studied.

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References:

[1] M O Nutt, J B Hughes and M S Wong, Environmental Science & Technology 39 (2005), p. 1346

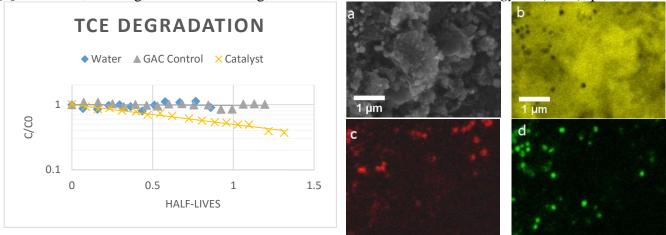


Figure 1. Batch test results in a pseudo-first order kinetics for degradation of trichloroethylene.

Figure 2. SEM images of Pd-Au-GAC catalysts (a), EDS map of carbon (b), Pd (c), and Au (d).

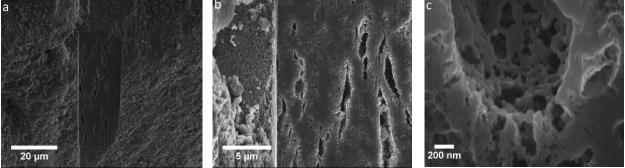


Figure 3. FIB across section of a GAC particle (a) at 3500X magnification, (b) at 15000X magnification and (c) at 150000X magnification, showing the inside of a pore of the GAC particle.

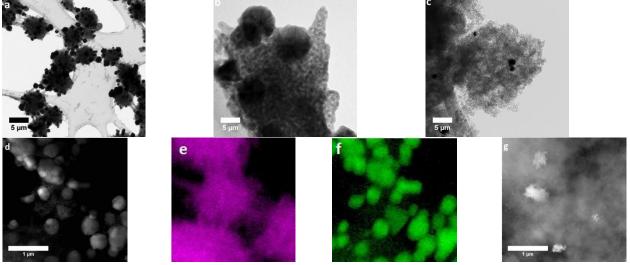


Figure 4. TEM images of Pd and Au NPs (a), High magnification of NPs (b), NPs on GAC (c), Dark field STEM image of NPs (d), EDS map of Pd (e), Au (f), Dark field STEM image of NPs on GAC (g).