Nitrate Photoremoval in Water Using Nanocatalysts Based on Ag / Pt over TiO2

Authors : Ana M. Antolín, Sandra Contreras, Francesc Medina, Didier Tichit

Abstract : Introduction: High levels of nitrates (> 50 ppm NO3-) in drinking water are potentially risky to human health. In the recent years, the trend of nitrate concentration in groundwater is rising in the EU and other countries. Conventional catalytic nitrate reduction processes into N2 and H2O lead to some toxic intermediates and by-products, such as NO2-, NH4+, and NOx gases. Alternatively, photocatalytic nitrate removal using solar irradiation and heterogeneous catalysts is a very promising and ecofriendly technique. It has been scarcely performed and more research on highly efficient catalysts is still needed. In this work, different nanocatalysts supported on Aeroxide Titania P25 (P25) have been prepared varying: 0.5-4 % wt. Ag); Pt (2, 4 % wt.); Pt precursor (H2PtCl6/K2PtCl6); and impregnation order of both metals. Pt was chosen in order to increase the selectivity to N2 and decrease that to NO2-. Catalysts were characterized by nitrogen physisorption, X-Ray diffraction, UV-visible spectroscopy, TEM and X Ray-Photoelectron Spectroscopy. The aim was to determine the influence of the composition and the preparation method of the catalysts on the conversion and selectivity in the nitrate reduction, as well as going through an overall and better understanding of the process. Nanocatalysts synthesis: For the mono and bimetallic catalysts preparation, wise-drop wetness impregnation of the precursors (AqNO3, H2PtCl6, K2PtCl6) followed by a reduction step (NaBH4) was used to obtain the metal colloids. Results and conclusions: Denitration experiments were performed in a 350 mL PTFE batch reactor under inert standard operational conditions, ultraviolet irradiations (λ =254 nm (UV-C); λ =365 nm (UV-A)), and presence/absence of hydrogen gas as a reducing agent, contrary to most studies using oxalic or formic acid. Samples were analyzed by Ionic Chromatography. Blank experiments using respectively P25 (dark conditions), hydrogen only and UV irradiations without hydrogen demonstrated a clear influence of the presence of hydrogen on nitrate reduction. Also, they demonstrated that UV irradiation increased the selectivity to N2. Interestingly, the best activity was obtained under ultraviolet lamps, especially at a closer wavelength to visible light irradiation ($\lambda = 365$ nm) and H2. 2% Ag/P25 leaded to the highest NO3conversion among the monometallic catalysts. However, nitrite quantities have to be diminished. On the other hand, practically no nitrate conversion was observed with the monometallics based on Pt/P25. Therefore, the amount of 2% Ag was chosen for the bimetallic catalysts. Regarding the bimetallic catalysts, it is observed that the metal impregnation order, amount and Pt precursor highly affects the results. Higher selectivity to the desirable N2 gas is obtained when Pt was firstly added, especially with K2PtCl6 as Pt precursor. This suggests that when Pt is secondly added, it covers the Ag particles, which are the most active in this reaction. It could be concluded that Ag allows the nitrate reduction step to nitrite, and Pt the nitrite reduction step toward the desirable N2 gas.

Keywords : heterogeneous catalysis, hydrogenation, nanocatalyst, nitrate removal, photocatalysis

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