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Materials For Energy Storage For Around-the-clock Photocatalysis

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Photocatalysis is emerging as a leading approach to the destruction of harmful chemical and biological threats at ambient temperatures, ideally using only the energy of the sun. Naturally, this process is effective only during the day, necessitating the development of strategies that extend the operational validity of photocatalytic materials to low-light or night-time conditions. Various materials have been developed over the last two decades towards this goal, typically by creating materials combinations that can both absorb light and store the photogenerated charge carriers, for example through 'spill-over' of photoexcited electrons from the absorbing component to the storage one. The capacity of a material to store charges depends on a range of physicochemical features, including the crystallographic nature of materials' interfaces, which we investigate here. We implemented a model system consisting of gold nanoparticles (AuNPs) supported on titanium dioxide (TiO2) anatase nanocrystals with predominantly (101), (100), or (001) facets. Cyclic voltammetry in dark, anaerobic conditions showed that all three materials exhibited increased current densities with increasing illumination time, with the highest increase observed for Au/TiO2(001). We further employed photocharged Au/TiO2 particles for catalytic reactions in the dark and found a consistent trend of Au/TiO2(001) being the most active. Using density functional theory, we calculated the Bader charge and the partial density of states, revealing that the presence of additional oxygen atoms at the Au/TiO2 interface leads to charge depletion from Au, providing more accessible vacant states to accept electrons from TiO2, with calculations showing the greatest charge depletion for Au/TiO2(001). Our results suggest that crystal surface engineering can be used as a powerful tool to optimize materials for electron storage.

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