Clay-Based Interfacial Bond Engineering: Clays as Charge-Transfer Mediators for Photo/Piezocatalytic Hydrogen Production

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Clay minerals are natural or synthetic nanostructured silicates composed of tetrahedral SiO_4 sheets combined with octahedral layers containing Al, Mg, or Fe. Their high surface area, ion-exchange capacity, surface hydroxyl groups, tunable charge, adsorption ability, and chemical stability make them more than passive catalyst supports. In catalytic systems, clays can limit nanoparticle agglomeration, concentrate reactants near active sites, stabilize dispersed phases, and mediate charge migration across solid–solid interfaces. Their use first expanded in photocatalysis, where clay-supported TiO_2, ZnO, g-C_3N_4, and metal sulfides were applied mainly to pollutant degradation and later to CO_2 conversion and hydrogen evolution. More recently, clay-containing systems have attracted attention in piezocatalysis, in which mechanical deformation or ultrasonication induces polarization charges in non-centrosymmetric semiconductors. Although most clays are not intrinsically piezoactive, they can improve piezo- and photopiezocatalytic performance by enhancing catalyst dispersion, shortening carrier-transport distances, stabilizing active domains, and facilitating interfacial electron transfer. Surface hydroxyl groups can form Si–O–M or Al–O–M linkages, converting simple physical contact into chemically bridged charge-transfer pathways. Such interfacial bond engineering can suppress charge recombination and accelerate directional electron migration. Clay morphology is particularly important. Halloysite offers a tubular architecture with chemically distinct inner and outer surfaces, sepiolite provides fibrous channels and high-aspect-ratio transport pathways, while Laponite consists of nanoscale disks that create extensive two-dimensional contact with semiconductor particles. These differences influence catalyst nucleation, particle size, interfacial area, and electron-transport behaviour. In Laponite/ZnO systems, Si–O–Zn bonds and finely dispersed ZnO nanoparticles demonstrate how morphology and bond chemistry can act cooperatively. Overall, comparing halloysite, Laponite, and sepiolite provides a framework for designing low-cost, stable, and scalable clay-based interfaces for photo/piezocatalytic hydrogen production.

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