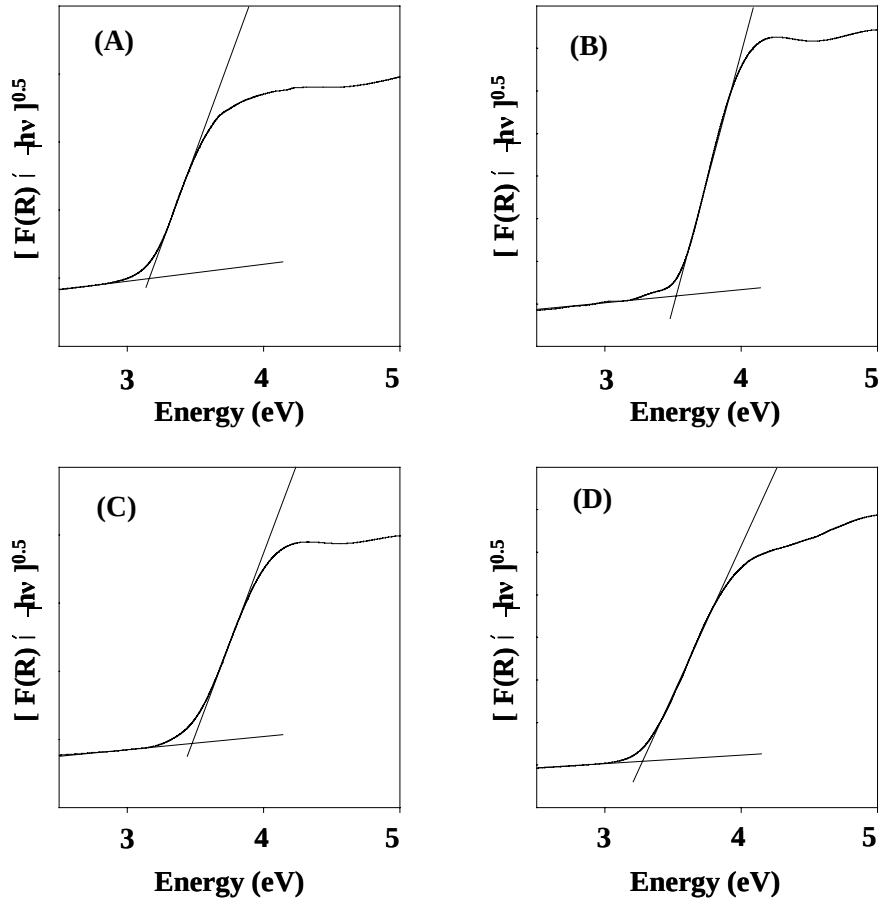
Supporting Information

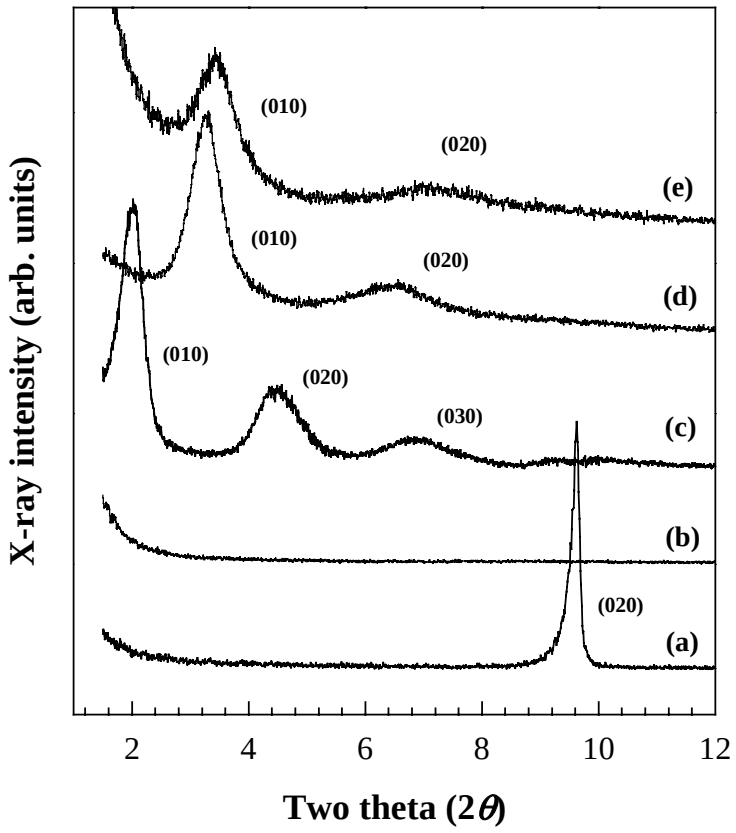
Synthesis of microporous hybrid. The host layer compound, cesium titanate ($Cs_xTi_{2-x/4}\square_{x/4}O_4$, x = 0.67), was prepared by the solid-state reaction of a stoichiometric mixture of Cs_2CO_3 and TiO_2 at 800 ºC for 20 h. The protonic form ($H_xTi_{2-x/4}\square_{x/4}O_4{\cdot}H_2O$, x = 0.67) was obtained by stirring the cesium titanate in a HCl solution (1 M) at room temperature for 3 days. The HCl solution was replaced with a fresh one every 24 h. The layered protonic titanate (1 g) was exfoliated into titanate single layers by intercalating 250 ml of the TBAOH solution (0.016 M) for 7 days at room temperature. The TBA/H molar ratio was adjusted to cation-exchange capacity (CEC) of the layered protonic titanate, which is a favorable condition for exfoliation.

Titanium isopropoxide (30 ml) with acetylacetone (20.4 ml) was added dropwise to vigorously stirred mixture solution of 180 ml of distilled water and 2 ml of HNO_3 (14 M). The resulting solution was peptized by further stirring at 60 °C for 8 h, which gives rise to a mono-dispersed and non-aggregated TiO_2 nano-sol with anatase phase.

A TiO_2-pillared layered titanate has been prepared by hybridizing the exfoliated layered titanate solution (20 ml) with the anatase TiO_2 nano-sol (20 ml) at 60 °C for 24 h. The product was collected by centrifuging (12000 rpm, 10 min), washed with a mixed solution of distilled water and ethanol (1:1, v/v) to remove excess TiO_2 sol, and dried in ambient atmosphere. Finally, the obtained material was heated at 300 °C for 2 h in order to complete the pillaring process.



S1. Plots of [F(R) × hν]$^{0.5}$ vs energy, where R is the reflectance from the UV-vis diffuse reflectance measurements: (A) TiO_2 xerogel, E_g = 3.2 eV: (B) layered cesium titanate, E_g = 3.5 eV: (C) layered protonic titanate, E_g = 3.4 eV: (D) nanohybrid-I, E_g = 3.3 eV. The intercept of the two linearly extrapolated lines gives the bandgap energy (E_g).



S2. Powder XRD patterns for microporous nanohybrid derivatives: (a) layered titanate, (b) the colloidal suspension of exfoliated titanate, (c) as-prepared microporous nanohybrid, (d) microporous nanohybrid with the heat-treatment at 300 ^{o}C, (e) microporous nanohybrid with the heat-treatment at 350 ^{o}C.