Supplementary information

Experimental section

The isoelectric point was calculated from zeta potential measurements at different pH values. Laser Doppler velocimetry was applied to characterize the electrophoretic mobility (EPM) of the particles using a Malvern Zetasizer Nano ZS. Measured EPMs were converted to zeta potential using the Smoluchowski equation. Disposable folded capillary cells were employed. First, 0.05 g of solid was suspended in 15 mL of Milli-Q water and the solution pH was modified by adding HCl or NaOH.

Sample	pH_{PZC}
@TiO_2TPA00$_{T100}$	6.1
@TiO_2TPA10$_{T100}$	5.1
@TiO_2TPA20$_{T100}$	4.7
@TiO_2TPA30$_{T100}$	3.9
@TiO_2TPA00$_{T500}$	5.9
@TiO_2TPA10$_{T100}$	4.8
@TiO_2TPA20$_{T100}$	3.9
@TiO_2TPA30$_{T100}$	2.7

Synthesis of SiO_2@TiO_2

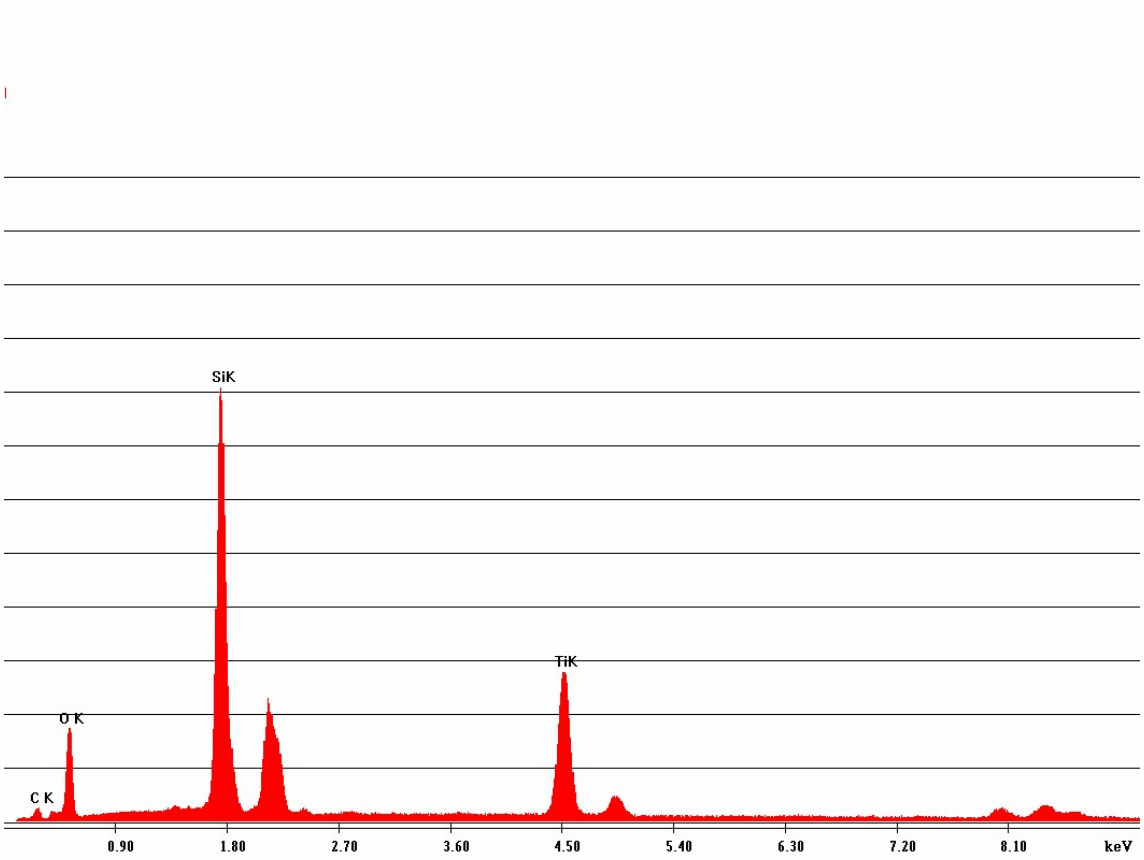
Spherical particles with an amorphous, solid core of SiO_2 and a porous, polycrystalline shell of TiO_2 formed in a three-step procedure. The first step yielded silica spheres, the second step produced sphere of silica covered with a layer of amorphous titanium oxide, and the last step crystallized the titanium oxide in the shell with a mild chemical treatment. Synthesis of silica spheres with a narrow distribution in size followed as described in an earlier contribution [S1]. Formation of a homogenously thick layer of titanium oxide followed likewise as previously described [S2]. We adapted a procedure developed for amorphous gels of TiO_2 to crystallize the shell [S3]. Thus, we obtained core@shell spheres with an amorphous, solid core of SiO_2 and a crystalline, porous shell of TiO_2 (SiO_2@TiO_2).
The shell crystallized with a mild chemical treatment. First, we poured 16.62 g of ethanol and then 0.083 g of HCl 36.5–38.0% from a glass beaker into a clean round-bottom flask (200 mL, one neck) in a fume hood and closed the flask with a septum. Then, we added 0.500 ± 0.001 g of silica spheres covered with amorphous TiO_2, a magnetic stir bar, and closed again the flask with the septum. We immersed the closed flask in an oil bath until the liquid surface in the flask remained below the liquid surface of the oil. After inserting a metallic needle (outer diameter 2 mm) through the septum, we turned on the heating plate. The temperature in the oil bath rose to 100.0 ± 0.1 °C under slow magnetic stirring. After reaching 100.0 ± 0.1 °C, the flask remained at this temperature 12 h. During this time, the liquid evaporated through the needle and a pale yellow solid appeared inside the flask. Then, we removed the flask from the oil bath and let it cool down to room temperature. After opening the flask, we removed the solid inside from the inner glass wall with a metallic spatula, collected it over a paper sheet, and finally swept it into a glass vial, which we closed with a screw lid.

Synthesis of @TiO2

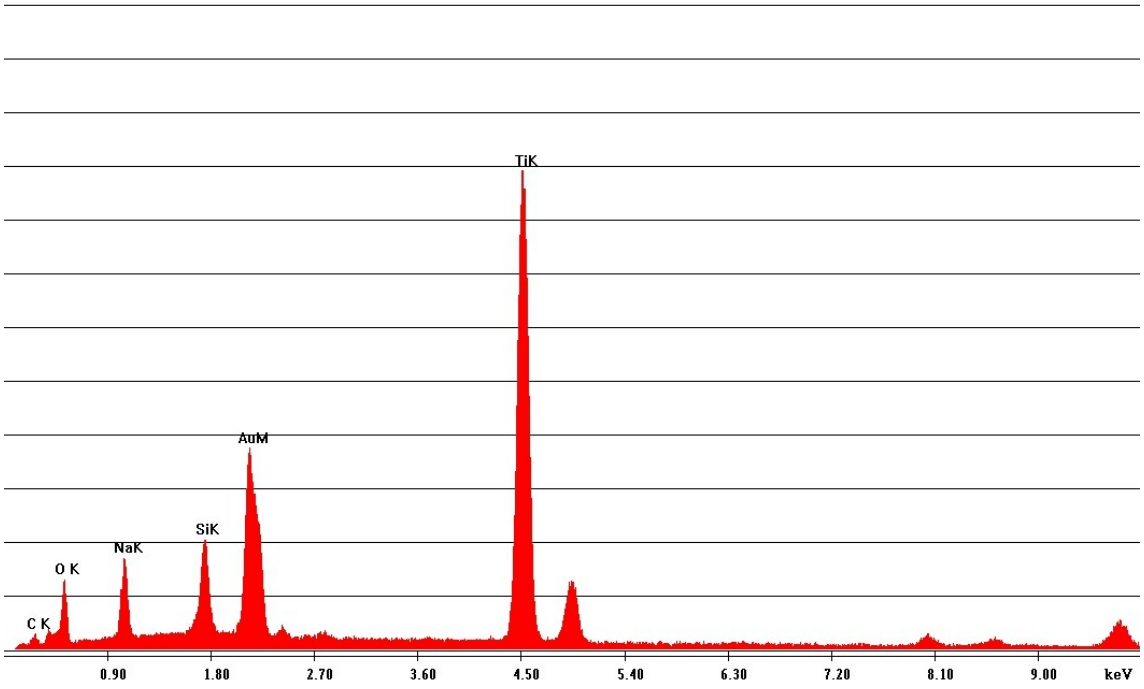
In the final step, the silica from the composite particles was removed by a treatment with sodium hydroxide solution (1 N). First, all samples were kept for 16 h at 30 °C in the solution.

After that, the alkaline solution was changed for a fresh one, and the extraction was continued for the same time at 30 °C. To completely remove the sodium hydroxide solution, the hollow particles were suction filtered, washed five times with H_2O and three times with ethanol (50 mL in each wash).

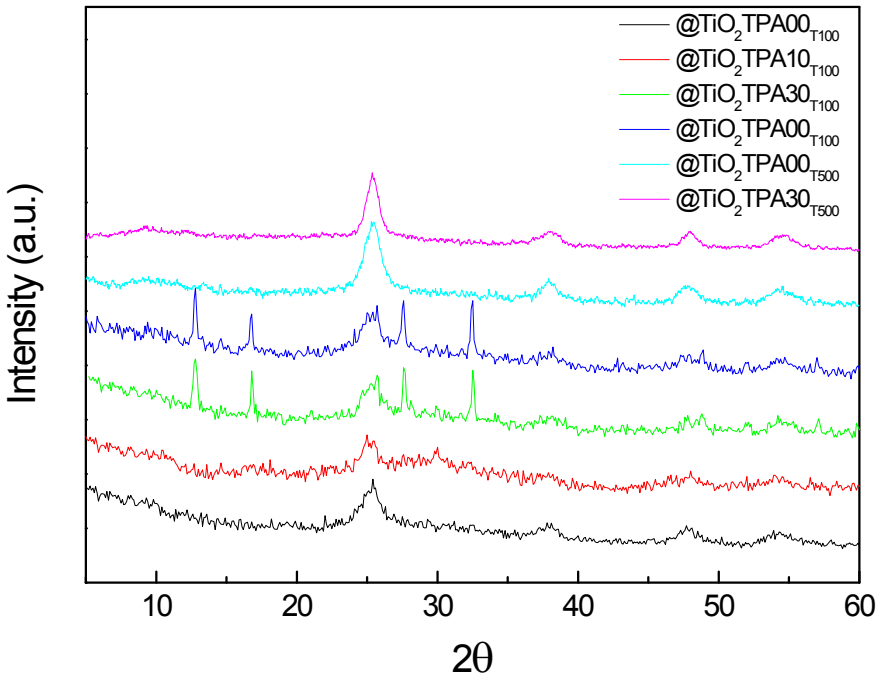
[S1] P.M. Arnal, C. Weidenthaler, F. Schueth, Chem. Mater. 18 (2006) 2733.
[S2] P.M. Arnal, The Synthesis of Monodisperse Colloidal Core@Shell Spheres and Hollow Particles, Ruhr Universität Bochum, Bochum, Doktor der Naturwissenschaften, 2006.
[S3] D.P. Serrano, G. Calleja, R. Sanz, P. Pizarro, J. Mater. Chem. 17 (2007) 1178.



SIFigure 1. EDX patterns of SiO_2@TiO_2 sample.



SIFigure 2. EDX patterns of @TiO_2 sample.



SIFigure 3. XRD patterns of @TiO2TPAXX$_{T100}$, @TiO2TPA00$_{T500}$ and @TiO2TPA30$_{T500}$ samples.