Supporting Information:

**Figure S1.** Changes of TOC over the course of the photocatalytic degradation of AO7 (2.0 x 10⁻⁵ M, 30 ml) in the presence of the CTF-M (20 mg) under exposure to UV light.

**Figure S2.** Absorption spectra of AO7 (2.0 x 10⁻⁵ M, 30 ml) solutions in the presence of CTF-M (20 mg) without UV light irradiation. These data are for the adsorption test of CTF-M with the same experimental conditions as Figure 6, indicating the color removal rate without UV light irradiation is almost one time slower than that with UV light irradiation.

**Figure S3.** Changes of TOC without UV light irradiation. These data are for the adsorption test of CTF-M with the same experimental conditions as Figure S1, indicating the TOC removal rate without UV light irradiation is almost one time slower than that with UV light irradiation.
Figure S4. Reusability of CTF-M measured by adsorption capacity for 10 consecutive cycles.

Detailed Experimental Section:
Preparation of chitosan solution
Deionised water (99.3 ml) was mixed with 0.7 ml of acetic acid in a beaker using a magnetic stirrer. Subsequently chitosan powder (Harvard Biotech, MA, USA, molecular weight 731 g/mol) was added to this mixture gradually in order to prevent agglomeration. The amount of chitosan added to the mixture was 2 g resulting in concentrations of 2.0 wt%. This process was performed at ambient temperature (22 °C). Once the suspension had become clear in appearance, indicating the total dissolution of the chitosan.

Preparation of Fe3O4 nanoparticles
First, 10.0 mL of FeSO4 solutions ([Fe2+], 0.25 M) and 10.0 mL of Fe2(SO4)3 solutions ([Fe3+], 0.25 M) were mixed. A solution of 5.0 M NaOH was then dropped slowly into the mixed solutions under stirring at room temperature until pH 12 was attained. The solutions were allowed to stand at room temperature and washed with distilled water until neutrality was obtained. The as-prepared Fe3O4 nanoparticles were collected by a magnet.

CTF microspheres preparation
10 ml of chitosan (2.0 wt%) in acetic acid was mixed with 0.05g Fe3O4 nanoparticles and 0.05g Degussa P25 TiO2 nanoparticles in a beaker under ultrasonic. This resulting precursor for CTF-M was drawn into a 10 ml syringe and perfused through a steel capillary of 0.7 mm internal diameter using a Harvard syringe pump (PHD-4400) and subjected to electrohydrodynamic atomisation (Fig. 1). The solution was pumped at a flow rate of 100 μl/min. The voltage used was 9 kV. Ion induced gelation was used as the primary mechanism of preparing the chitosan microspheres. 80 ml of distilled water with 0.1 g of NaOH was used as the collection solution, which could instantly gel the droplets of electrosprayed chitosan. The chitosan droplets in the collection solution kept spherical shape during the process. The as-prepared CTF-M microspheres were collected by a magnet. The samples were freeze-dried or heat-dried for storage.

The selection criteria for the amount of each component (porous chitosan/TiO2/Fe3O4):
1) for Fe3O4 which is in charge of magnetic collection, the amount we added is just enough to move CTF-M under an external field; it will offset the other functions if the amount of Fe3O4 is too much; for chitosan which is both in charge of adsorption and scaffold for the microspheres, the amount we added depends on how big the size of microspheres we want, because the chitosan polymer is viscous in the spray precursor, the amount of chitosan is the main factor to effect the microsphere size; for TiO2 which is in charge of photodegradation of pollutants, the amount we added is as much as possible, as long as the microspheres can keep the shape.

Adsorption capacity of CTF-M
The adsorption capacity of CTF-M was characterised by their AO7 dye uptakes. 10 mg of CTF-M was added to 100 mL of AO7 with concentration of 0.2 g/L in distilled water. The suspensions were kept in a shaker at room temperature during 24 h, which is enough for guaranteeing adsorption equilibrium of AO7 over CTF-M surfaces. The CTF-M suspensions were next collected by a magnet, and the residual AO7 contained in the solution was quantified by UV–Vis spectrophotometry at a fixed wavelength of 486 nm.
Photocatalytic activity of CTF-M
20 mg of CTF-M was added to 30 mL of AO7 with concentration of $2.0 \times 10^{-5}$ M in distilled water. The CTF-M suspensions were stirred under UV light (365 nm) and the residual AO7 in the solution was tested by UV–Vis spectrophotometry.

Reusability test of CTF-M
After the photodegradation of AO7, firstly, the CTF-M were collected by an external magnet; then the collected CTF-M were dried at 80 °C for 24 hours for reuse.

Characterization Methods
Surface morphologies were examined with a ZEISS EVO 50 scanning electron microscope (SEM). The optical photos were taken with an optical microscope (OLYMPUS SZX9). UV–vis measurements were taken with a Shimadzu UV-1700 spectrophotometer. TOC was measured by means of a Tekmar Dohrmann Apollo 9000 TOC analyzer. The structure was analyzed by X-ray diffraction (Bruker AXS Advance) with graphite-monochromatized Cu Ka radiation. Functional groups were obtained on Fourier transform infrared (FTIR) spectrometer (PERKin ELMER, Spectrum GX). The magnetic properties of was determined with a Vibrating Sample Magnetometer (Lakeshore 7300) (1.5 Tesla) with an applied field of between -5000 and 5000 Oe. Christ Freeze Dryer Alpha 1-2 was used for drying samples.