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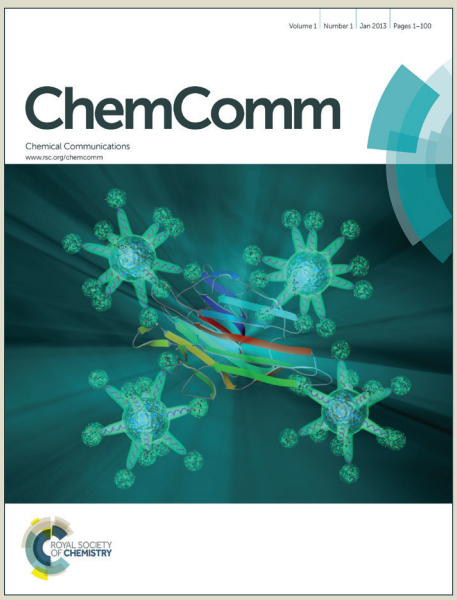

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Supporting Information

General techniques: Powder X-ray diffraction (PXRD) and thermogravimetric analysis (TGA), elemental analysis, transmission electron microscopy (TEM), N_2 adsorption-desorption, UV-visible (UV-vis) and fluorescence spectroscopy were employed to characterize the synthesized materials. PXRD measurements were performed on a D8 Advance diffractometer using Cu Kα radiation (Philips, Amsterdam, The Netherlands). Thermogravimetric analyses were carried out on a TGA/SDTA 851e balance (Mettler Toledo, Columbus, OH, USA), using an oxidizing atmosphere (air, 80 mL min^{-1}) with a heating program: gradient of 393-1273 K at 10°C min^{-1}, followed by an isothermal heating step at 1273°C for 30 min. TEM images were obtained with a 100 kV CM10 microscope (Philips). N_2 adsorption-desorption isotherms were recorded with an ASAP2010 automated sorption analyzer (Micromeritics, Norcross, GA, USA). The samples were degassed at 120°C in vacuo overnight. The specific surface areas were calculated from the adsorption data in the low pressure range using the Brunauer, Emmett and Teller (BET) model. Pore size was determined following the Barret, Joyner and Halenda (BJH) method. Fluorescence spectroscopy measurements were carried out on a Felix 32 Analysis version 1.2 (Build 56, Photon Technology International, Birmingham, NJ, USA).

Chemicals: Tetraethylorthosilicate (TEOS), *n*-cetyltrimethylammoniumbromide (CTAB), NaOH, **(**3-isocyanatopropyl)triethoxysilane, rhodamine B, tris(hydroxymethyl)aminomethane (TRIS), hydrochloric acid and oligonucleotides **O1** (NH_2-$(CH_2)_6$-5'-GACTACCAGGGTATC-3') and **O2** (5'-AAGCGTGGGGAGCAAACAGGATTAGATACCCTGGTAGTC-3') were purchased from Sigma-Aldrich Química (Madrid, Spain). All products were used as received.

Synthesis of MCM-41 mesoporous nanoparticles: NaOH (2.00 mol L^{-1}, 3.5 mL) was added to a solution of CTAB (1.00 g, 2.74 mmol) in deionized H_2O (480 mL). The solution temperature was

adjusted to 80°C and then TEOS (5.00 mL, 2.57×10^{-2} mol) was added dropwise. The mixture was stirred for 2 h to give a white precipitate. The solid was isolated by centrifugation and washed with deionized H_2O and EtOH and then dried at 60°C for 12 h to give MCM-41. In order to remove the template phase, MCM-41 was calcined at 550°C using an oxidizing atmosphere.

Synthesis of S1-I: In a typical synthesis, calcined MCM-41 (200 mg) and rhodamine B (766.4 mg, 0.16 mmol) were suspended in CH_3CN (10 mL). The suspension was stirred at room temperature for 24 h. Then an excess of (3-isocyanatopropyl)triethoxysilane (247.6 µl , 1.0 mmol) was added, and the final mixture was stirred at room temperature for 5.5 h. The resulting pink solid (**S1-I**) was isolated by centrifugation, washed with CH_3CN (5 mL) and dried at 38°C for 18 h.

Synthesis of S1-O1/O2: A mixture of oligonucleotides **O1** and **O2** (25 µL of each in water at 100 µM concentration) was heated to 90ºC and cooled at 25ºC slowly in order to achieve DNA hybridization (formation of **O1/O2**). The DNA mixture solution was added to a suspension containing 1 mg of **S1-I**, 50 µL of water, 700 µL of a solution of rhodamine B in CH_3CN (1 mM) and 2 µL of triethylamine. Finally, the mixture was stirred for 18 h at 37ºC. The resulting solid was isolated by centrifugation and washed with hybridization buffer (20 mM TRIS-HCl, 37.5 mM $MgCl_2$, pH 7.5) to eliminate the residual dye and unbounded oligonucleotides (see Figure S1).

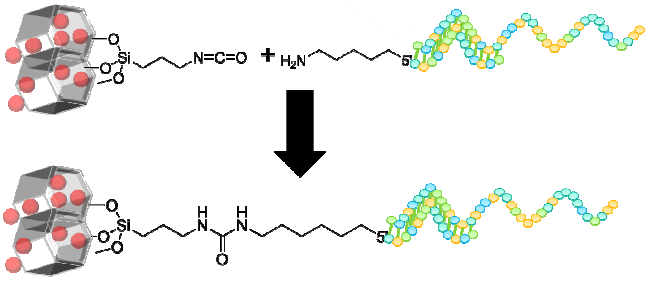

Figure S1. Surface functionalization reaction of **S1-I** via urea bond formation with **O1/O2**.

Characterization of the prepared materials: The MCM-41 scaffold and the mesoporous solid **S1-I** were characterized following standard techniques, including transmission electron microscopy (TEM), powder X-ray diffraction (PXRD) and N_2 sorption analysis**.** PXRD pattern of siliceous MCM-41 nanoparticles as synthesized (Figure S2, curve a) showed four low-angle reflections typical of a hexagonal array, indexed as (100), (110), (200), and (210) Bragg peaks. A significant displacement of 3Å of the (100) peak in the PXRD pattern of the MCM-41 calcined nanoparticles was evident in curve b, related to further condensation of silanol groups during the calcination step. Finally, curve c corresponds to the **S1-I** XRD pattern. An intensity decrease of the (100) peak and a broadening of the (110) and (200) reflections was observed, related to a loss of contrast due to the filling of the pore voids with the rhodamine B dye.

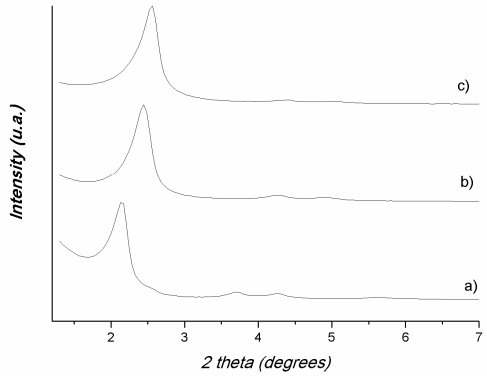

Figure S2. Powder X-ray patterns of the solids (a) MCM-41 as synthesized, (b) calcined MCM-41, and (c) solid **S1-I**.

The presence, in the final functionalized solids, of the mesoporous structure was confirmed by TEM analysis, in which the typical channels of the MCM-41 matrix are visualized as alternate black and white stripes (see Figure S3). The figure also shows that the prepared MCM-41 and solid **S1-I** are obtained as spherical particles with diameters ranging from ca. 80 to 120 nm.

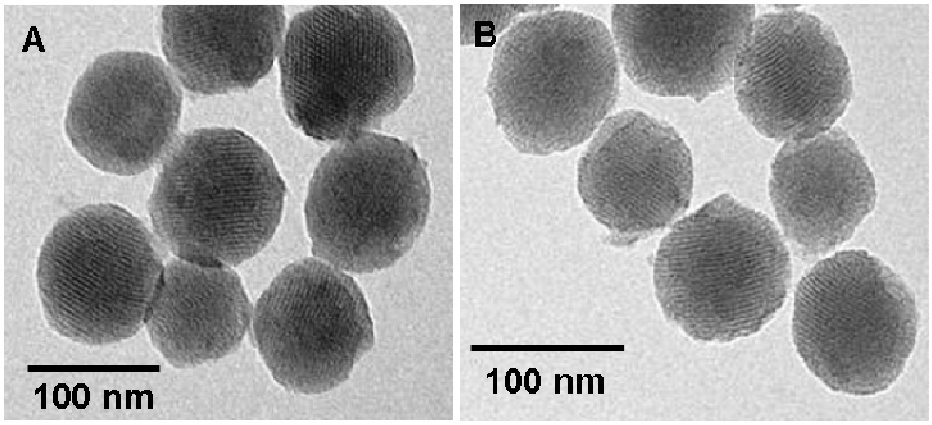

Figure S3. TEM images of calcined MCM-41 sample (A) and solid **S1-I** (B) showing the typical porosity of the MCM-41 mesoporous nanoparticles matrix.

The N_2 adsorption–desorption isotherms of the calcined MCM-41 nanoparticles show an adsorption step at an intermediate P/P_0 value (0.1–0.3) typical of this type of solid (see Figure S4, curve a). A total pore volume of 0.93 $cm^3\ g^{-1}$ was calculated by using the BJH model on the adsorption branch of the isotherm. The application of the BET model resulted in a value for the total specific surface of 1228.2 $m^2\ g^{-1}$. From the XRD, porosimetry, and TEM studies a pore diameter of 2.40 nm was determined. The N_2 adsorption–desorption isotherm of **S1-I** (see Figure S4, curve b) is typical of mesoporous systems with filled mesopores, and a significant decrease in the N_2 volume adsorbed (0.34 $cm^3\ g^{-1}$) and surface area (524.9 $m^2\ g^{-1}$) were observed.

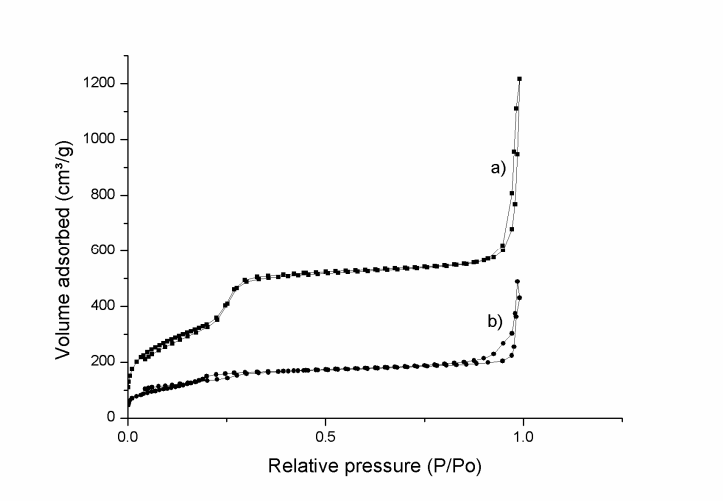

Figure S4. N_2 adsorption-desorption isotherms for calcined MCM-41 nanoparticles (a) and **S1-I** material (b).

The content of **(**3-isocyanatopropyl)triethoxysilane and rhodamine B in the prepared solid **S1-I** was determined by elemental and thermogravimetric analyses (see Table S1). Moreover the diameter of MCM-41, **S1-I** and **S1-O1/O2** solids was measured by dynamic light scattering (DLS) studies (see Figure S5 and Table S2).

Table S1. Contents of **(**3-isocyanatopropyl)triethoxysilane and rhodamine B (in mmol/g solid) in the prepared solid **S1-I**.

	$\alpha_{isocyanate}$ (%)	$\alpha_{Rhodamine\ B}$ (%)	$\alpha_{Organic\ matter}$ (%)
Elemental analysis	3.99±0.09	17.16±0.11	21.15±0.10
Thermogravimetry	-	-	26.1±0.2

Table S2. Diameter of MCM-41, **S1-I** and **S1-O1/O2** solids measured by DLS.

	Particle diameter (nm)
MCM-41	88±2
S1-I	114±3
S1-O1/O2	154±2

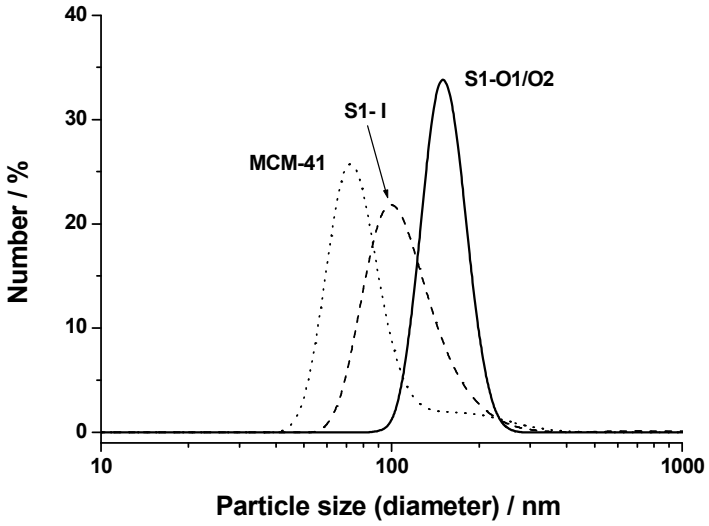

Figure S5. Size distribution by number of particles obtained by DLS studies for calcined MCM-41, **S1-I** and **S1-O1/O2** solids.

Controlled release of solid S1-O1/O2 in the presence of urease and DNAse I enzymes: To confirm the proper anchoring of the double stranded **O1/O2** oligonucleotide, 200 μg of solid **S1-O1/O2** were suspended in 400 μL of the hybridization buffer and separated in 2 aliquots (each of 200 μL). One of the samples was filled to a final volume of 1000 μL with hybridization buffer whereas the other was filled with the same buffer but containing DNAse I enzyme (at a concentration of 10 U/μL). Both suspensions were stirred at 25ºC for 90 minutes. Aliquots of 150 μL were taken at several times and centrifuged 2 min at 12000 rpm (in order to remove the solid) and the fluorescence of the rhodamine B released measured at 575 nm (λ_{exc} 555 nm). The same experimental procedure was used to study rhodamine B release form solid **S1-O1/O2** in the presence of urease. The obtained results are shown in Figure S6.

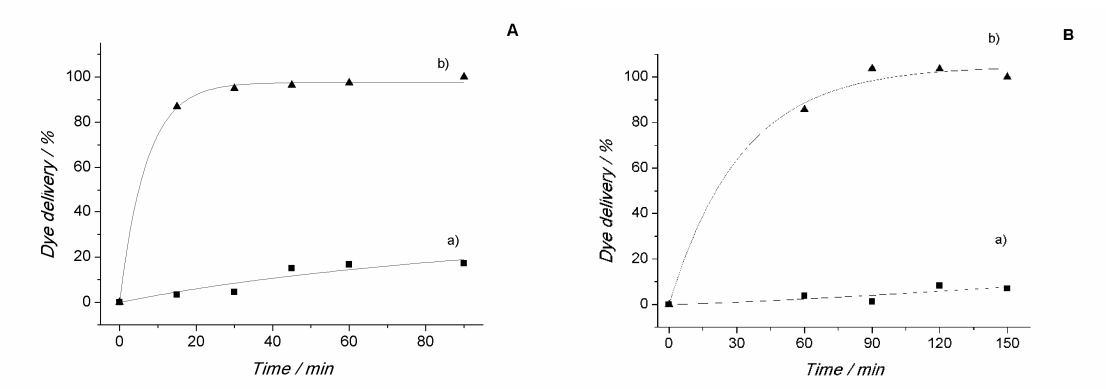

Figure S6. A: Delivery of rhodamine B from solid **S1-O1/O2** in the absence (a) and in the presence (b) of DNAse I; B: Delivery of rhodamine B from solid **S1-O1/O2** in the absence (a) and in the presence (b) of urease.

Release experiments of S1-O1/O2: To investigate the gating properties of **S1-O1/O2**, 200 μg of this solid were suspended in 400 μL of the hybridization buffer and separated in 2 aliquots of 200 μL. Both samples were filled to a volume of 900 μL with hybridization buffer. At the same time a solution of 4000 copies μL^{-1} of *Mycoplasma fermentans* quantification standard in water was heated to 95ºC for 5 min and cooled on ice bath at 0ºC for 3 min (in order to dehybridize the double stranded structure). Then, 100 μL of the genome solution was added to one of the **S1-O1/O2** suspensions. Simultaneously 100 μL of water (milliQ grade) with the same thermal treatment were added to the remaining aliquot. Both suspensions were stirred at 25ºC for 60 min. Aliquots of 150 μL were taken at several times and centrifuged 2 min at 12000 rpm (in order to remove the solid) and the fluorescence of the rhodamine B released measured at 575 nm (λ_{exc} 555 nm).

Calibration curve of S1-O1/O2 with *Mycoplasma fermentans* genomic DNA: In order to carry out these experiments, 120 μL of aqueous solutions containing different number of *Mycoplasma fermentans* DNA copies were heated for 5 min at 95ºC, in order to dehybridize DNA. Then, these solutions were left cooling for ca. 3 min in ice bath. These samples were added to several aliquots of 60 μL of **S1-O1/O2** (0.2 mg of solid in 400 μL of hybridization buffer). All samples were stirred at 25ºC for 30 min, and after that, were centrifuged 2 min at 12000 rpm in order to remove the solid. The fluorescence of the released

rhodamine B was then measured at 575 nm (λ_{exc} 555 nm). The obtained calibration curve is shown in Figure S7. From this curve the limit of detection (ca. 70 DNA copies μL^{-1}) has been calculated based in the intersection point of the two slopes. Moreover a limit of quantification (10σ) of ca. 150 DNA copies μL^{-1} has been determined.

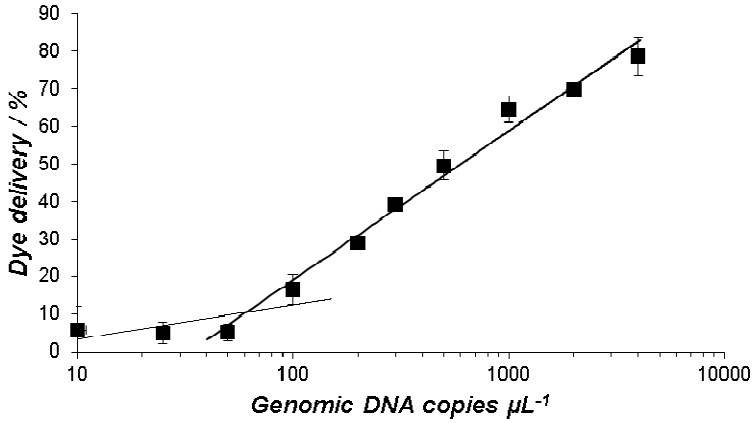

Figure S7. Release of rhodamine B from solid **S1-O1/O2** in the presence of different numbers of *Mycoplasma fermentans* genomic DNA copies in Tris-HCl buffer (pH 7.5).

Selectivity studies with S1-O1/O2: In order to carry out these experiments, 120 µL of aqueous solutions containing different genome solutions of *Mycoplasma fermentans, Candida albicans, Legionella pneumophilla,* a mixture of *Candida albicans-Legionella pneumophilla* and a mixture of the three genomes (1000 copies μL^{-1}) were heated for 5 min at 95ºC, in order to dehybridize DNA. As control a 120 µL of hybridization buffer received the same thermal treatment. Then, these samples were left cooling for ca. 3 min in ice bath. These samples were added to several aliquots of 60 µL of **S1-O1/O2** (0.2 mg of solid in 400 µL of hybridization buffer). All samples were stirred at 25ºC for 30 min, and after that, samples were centrifuged 2 min at 12000 rpm in order to remove the solid. The fluorescence of the released rhodamine B was then measured at 575 nm (λ_{exc} 555 nm).