

ELECTRONIC SUPPLEMENTARY MATERIAL

A comparative life cycle assessment of the synthesis of mesoporous silica materials at small and large scale.

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Table S1.: Life Cycle Inventory for the preparation of 1 kg of silica mesoporous material

Process	Unit	Scenarios	FDU-1	HMS	KIT-5	KIT-6	MCM-41	MCM-48	MSP	MSU	nano-MCM-41	nano-MCM-48	SBA-15	SBA-16	UVM-7
Acetone	kg	1	-	-	-	-	-	-	-	2,2	-	-	160	-	-
Ammonia	kg	1 to 3, 5, 6	-	0,3	-	-	3,3	-	-	-	-	-	-	-	-
Ammonium nitrate	kg	1	-	1,1	-	-	-	-	-	-	1,9	5,6	-	-	-
Ammonium nitrate	kg	2 to 7	-	1,6	-	-	-	-	-	-	0,5	5,6	-	-	-
B50-6600	Kg	1 to 7	0,8	-	-	-	-	-	-	-	-	-	-	-	-
Butanol	kg	1 to 7	-	-	-	1,6	-	-	-	-	-	-	-	2,5	-
CTABr	kg	1 to 7	-	0,6	-	-	0,8	3,3	3,2	-	0,7	1,0	-	-	1,5
Ethanol	kg	1	20	307	-	-	-	-	120	-	2.272	52	-	-	5,4
Ethanol	kg	2, 6	3,3	416	3,3	-	3,3	3,3	28	-	144	55	3,3	3,3	3,3
Ethanol	kg	3, 4	-	407	-	-	-	-	25	-	125	52	-	-	-
Ethanol	kg	5, 7	2,9	21	0,2	-	0,2	0,2	1,4	-	7,2	2,7	0,2	0,2	0,2
Hydrochloric acid	kg	1 to 3, 5, 6	3,2	-	0,6	1,4	-	-	-	-	-	-	10	3,6	-
Hydrochloric acid	kg	4, 7	3,2	0,5	0,6	1,4	-	-	-	1,0	-	-	10	3,6	-

Process	Unit	Scenarios	FDU-1	HMS	KIT-5	KIT-6	MCM-41	MCM-48	MSP	MSU	nano-MCM-41	nano-MCM-48	SBA-15	SBA-16	UVM-7
Pluronic F127	Kg	1 to 7	-	-	0,7	-	-	-	-	-	-	3,9	-	0,8	-
Pluronic P123	Kg	1 to 7	-	-	-	1,6	-	-	-	-	-	-	-	-	-
POL1	Kg	1 to 7	-	-	-	-	-	-	-	-	-	-	1,6	-	-
Potassium chloride	kg	1 to 7	-	-	-	-	-	-	-	-	-	-	-	2,1	-
PtBA latex	kg	1 to 7	-	1,2	-	-	-	-	-	-	-	-	-	-	-
Sodium fluoride	g	1 to 7	-	-	-	-	-	-	-	14,0	-	-	-	-	-
Sodium hydroxide	kg	1 to 3, 5, 6	-	-	-	-	-	0,6	-	-	0,8	-	-	-	-
Sodium silicate	kg	4, 7	1,4	1,4	1,4	1,4	1,4	1,4	1,4	1,4	1,4	1,4	1,4	1,4	1,4
Sulfuric acid	kg	1 to 7	-	-	-	-	-	-	-	-	-	-	176	-	-
Tergitol 15-S-12	kg	1 to 7	-	-	-	-	-	-	-	2,2	-	-	-	-	-
Tetraethyl orthosilicate	kg	1 to 3, 5, 6	3,5	3,5	3,5	3,5	3,5	3,5	3,5	3,5	3,5	3,5	3,5	3,5	3,5
Triethanolamine	kg	1 to 7	-	-	-	-	-	-	9,5	-	-	-	-	-	8,8
Water, deionised	kg	1	250	407	179	59	77	408	322	104	752	2.143	420	467	95
Water, deionised	kg	2, 5 to 7	67	474	101	59	108	103	208	104	438	287	79	117	94
Water, deionised	kg	3, 4	15	422	50	59	57	51	203	104	447	235	27	65	42
Electricity	MJ	1	48.305	15.696	11.369	8.910	5.656	19.026	3.280	53.359	2.369	37.843	21.193	10.066	6.365
Electricity	MJ	2	3.121	3.757	1.273	1.244	690	1.089	769	1.526	4.639	636	3.022	921	694
Electricity	MJ	3, 4	7	2	8	8	7	7	7	13	13	13	11	8	7
Electricity	MJ	5	3.122	4.587	1.279	1.244	697	1.096	825	1.526	4.927	746	3.029	928	700
Electricity (rebewable)	MJ	6	3.121	3.757	1.273	1.244	690	1.089	769	1.526	4.639	636	3.022	921	694
Electricity (rebewable)	MJ	7	3.122	4.587	1.279	1.244	697	1.096	825	1.526	4.927	746	3.029	928	700
Heat from gas	MJ	3, 4	37	25	7	11	-	-	10	18	214	0	17	9	10

Summary of the synthesis procedures of the mesoporous materials

Synthesis of MCM-41.

2.4 g n-Alkyltrimethylammonium bromide was dissolved in 120 g of deionized water to yield a 0.055 mol L⁻¹ solution, and 9.5 g of aqueous ammonia (25 wt.%, 0.14 mol) was added to the solution. While stirring 10 g of tetraethoxysilane (0.05 mol) was added slowly to the surfactant solution over a period of 15 min resulting in a gel with the following molar composition: 1 TEOS:0.152 n-alkyltrimethylammonium bromide:2.8 NH₃:141.2 H₂O The mixture was stirred for one hour, then the white precipitate was filtered and washed with 100 ml of deionized water. After drying at 363 K for 12 h, the sample was heated to 823 K (rate: 1 K min⁻¹) in air and kept at this temperature for 5 h to remove the template. (M. Grün, K.K. Unger, A. Matsumoto, K. Tsutsumi, *Microporous and Mesoporous Materials*, 1999, 27, 207.)

Synthesis of MCM-48.

10 mL of TEOS were added to an aqueous solution containing 88 g of CTAB solution (10 wt %) and 10 mL of 2 M NaOH. After being stirred for about 0.5 h, the resulting homogeneous mixture was crystallized under static hydrothermal conditions at 373 K in a Teflon bottle for 72 h. The molar composition of the initial gel mixture was 1:0.23:0.55:112 SiO₂/Na₂O/CTAB/H₂O. The product was filtered out, thoroughly washed with deionized water and air-dried overnight. Such as-synthesized samples were calcined at 823 K first in flowing nitrogen for 1 h and then in flowing oxygen for 14 h to remove the CTAB. (J. Xu, Z. Luan, H. He, W. Zhou, L. Kevan, *Chemistry of Materials*, 1998, 10, 3690.)

Synthesis of UVM-7

44 mL of TEOS was mixed with 100 mL of TEAH3 and heated up to 140 °C. The reaction was allowed to cool, adding 18.7 g of CTAB at 120 °C and 320 mL of water at 80 °C. A white suspension was formed and allowed to age for 24 h at room temperature. The solid was filtered, washed with distilled water, dried in the oven at 50 °C for one day and calcined at 550 °C to remove the surfactant. (S. Muñoz-Pina, P. Amorós, J. El Haskouri, A. Andrés, J.V. Ros-Lis, *Nanomaterials*, 2020, 10, 1927.)

Synthesis of mesoporous Stober particles

22 mL of TEOS were mixed with 50 mL of TEAH3 and heated up to 140 °C. The reaction was allowed to cool, adding 18.7 g of CTAB at 120 °C and a mixture of 500 mL of ethanol and 900 mL of water at 70 °C. A white suspension was formed and allowed to age for 24 h at room temperature. The solid was removed by centrifugation (7500 rpm), washed with water and ethanol twice, dried in the oven at 50 °C for 24 h and calcined at 550 °C for surfactant removal. (S. Muñoz-Pina, P. Amorós, J. El Haskouri, A. Andrés, J.V. Ros-Lis, *Nanomaterials*, 2020, 10, 1927.)

Synthesis of SBA-15

4 g of triblock copolymer, poly (ethylene glycol)-block-poly (propylene glycol)-block-poly (ethylene glycol) [EO20-PO70-EO20, template], was dispersed in 30 g distilled water and stirred for 1 h. To the resulting solution, 25 g of 37% HCl was added under stirring and finally 8.5 g of tetraethoxysilane (TEOS, a silicate precursor) was added. The mixture was stirred at 35 °C or 40 °C for 20 h, followed by aging at 90 °C or 100 °C for 24 h. The solid was filtered and dried at 80 °C overnight. For the treatment with sulfuric acid, 1.0 g of 'as-synthesized' SBA-15 was mixed with 100 mL of 48 wt% H₂SO₄ solution and heated at 95 °C for 20 h. After reaction, the products were washed with water until the eluent became neutral, then washed with acetone, and dried at 80 °C. To remove the occluded (ethylene glycol) EO chains in the silica walls, sulfuric acid treated samples were further heated for 3–6 h in air at 300 °C. For the calcinated samples, each

dried sample was calcinated at 550 °C at 10 °C/min rate and held at 550 °C for 6 h. (H. Zhao, H. Han, *Journal of Solid State Chemistry*, 2020, 282, 121074.)

Synthesis of SBA-16

2g of F127 (EO106PO70EO106) and 5 g of inorganic salt KCl were added in 120mL HCl (2mol/L), the mixture was transferred to a synthesis to control the synthesis temperature, and 6 g of co-surfactant butanol was added after stirring at 38 °C for 2h. 8.3 g of silicon source tetraethylorthosilicate (TEOS) was added after stirring for 10 min, then kept static for 24 h before transferring the mixture into autoclave at 100 °C for 24h. The solid was filtrated with deionized water and dried at 80 °C for 6 h. At last, the SBA-16 material was synthesized successfully after calcining at 550 °C for 6h. (Z. Cao, P. Du, A. Duan, R. Guo, Z. Zhao, H. Zhang, P. Zheng, C. Xu, Z. Chen, *Chemical Engineering Science*, 2016, 155, 141.)

Synthesis of HMS

0.16 g of CTAB was dispersed in a mixed solution of deionized water (10 ml), ethanol (5 ml) and aqueous ammonia solution (25 wt%, 0.37 ml). The mixture was stirred for 15 min followed by dripping off 2.2 ml of PtBA latex. Finally, 1 ml of tetraethyl orthosilicate (TEOS) was successively added to the mixture, and the reaction was carried out for 24 h. After washing twice with water and ethanol, the product was refluxed in the ethanol solution of NH₄NO₃ (10 mg ml⁻¹) at 70 °C for 5 h. The synthesized product was then centrifuged twice with ethanol. (Sh. Ghasemi, Z. Jomeh Farsangi, A. Beitollahi, M. Mirkazemi, S.M. Rezayat, S. Sarkar, *Ceramics International*, 2017, 43, 11225.)

Synthesis of KIT-5

2.5 g of pluronic F127 was dissolved in 120 ml of distilled water containing 5.25 ml of concentrated hydrochloric acid (35 wt% HCl). To this mixture, 12 g of TEOS was added. The mixture was stirred at 45 °C for 24 h for the formation of mesostructured product. After 24 h, the reaction mixture was kept in a Teflon lined autoclave and heated for 24 h at 150 °C. The solid product was then filtered, washed with deionized water and dried at 80 °C. Finally, the sample were calcined at 550 °C for 6 h in order to remove the template. (D.N. Peter, R. Pushpakumar, E. Jayaseelan, N. Ananthi, *Materials Today: Proceedings*, 2021, 47, 739.)

Synthesis of KIT-6

6 g amphiphilic triblock copolymer Pluronic P123 (Mw = 5800, EO20-PO70-EO20), 11.8 g hydrochloric acid and 6 g butanol were dissolved in 217 g of deionized water under stirring at 35 °C. After stirring for 4 h, 12.9 g tetraethyl orthosilicate (TEOS, C₈H₂₀O₄Si) was added into the above solution and it was stirred again at 35 °C for 24 h to ensure that the solution was distributed homogenously. The resulting solution was transferred into a Teflon-lined autoclave and kept at different temperatures for 24 h under static conditions. The product was filtered and dried at 100 °C. After that, the samples were further calcined at 550 °C for 5 h in air. (B. Zhou, C.Y. Li, N. Qi, M. Jiang, B. Wang, Z.Q. Chen, *Applied Surface Science*, 2018, 450, 31.)

Synthesis of MSU

1.028 g of Tergitol 15-S-12 was first dissolved in 50 mL of water. Once surfactant dissolution was complete, 1.664 g of TEOS was added dropwise under magnetic stirring, then sonication was performed in an ice-bath, with a 600 W tunable Vibracell 72412 (Bioblock Scientific) ultrasound gun (probe diameter, 10 mm) along with continuous magnetic stirring. The sonicator was set at a defined potential power but the power actually transmitted to the medium depended on its composition (the effective delivered value is measured by the sonicator). Sodium fluoride (0.84 mL of a 0.238 M NaF solution) was then added in order to reach the molar ratio NaF : TEOS ~ 2%. The solution thus obtained was aged under slow shaking at 35 C for 48 h. A white colloidal

suspension appeared that settled progressively. The powder was filtered, dried, and calcined in air at 200 °C for 6 h, then at 620 °C for 6 h (heating rate, 5 °C min⁻¹) (E. Prouzet, F. Cot, C. Boissière, P.J. Kooyman, A. Larbot, *Journal of Materials Chemistry*, 2002, 12, 1553)

Synthesis of FDU

0.50 g of B50-6600 was dissolved in 30 g of 2 mol/L HCl. To this homogeneous solution, 2.08 g (0.01 mol) of tetraethyloxysilane (TEOS) was added with vigorous stirring for 24 h (TEOS:B50-6600:HCl:H₂O 1:0.0074:6:166, molar ratio). The resulting solid was aged in 100 °C for another 24 h. The solid product was filtered, washed, and dried in vacuum at room temperature. The calcination was carried out in an oven by slowly increasing temperature from room temperature to 550 °C in 4 h and heated at 550 °C for 6 h in air. (C. Yu, Y. Yu, L. Miao, D. Zhao, *Microporous and Mesoporous Materials*, 2001, 44-45, 65.)

Synthetic of nano-MCM-41

5 mL TEOS were dropwise added (0.25 mL min⁻¹) over a thermally stabilized solution (80 °C) composed by 1 g CTAB, 3.5 mL of 2M NaOH (14 mM) in 500 mL of double distilled water (resulting pH = 11.8). Once the addition was concluded, the mixture was left to react for a certain time (up to 2 h) to allow full silica condensation. The obtained MSNs were isolated by centrifugation (10000 rpm, 10 min) and repeated washings with 500 mL water (2) and ethanol (2) to remove excess reagents. The surfactant contained within the pores was removed under very mild conditions by washing the recovered nanoparticles with a slightly acidic ion-exchange solution (2 x 2 h reflux cycles with 250 mL of a 10 g L⁻¹ solution of NH₄NO₃ in 95:5 ethanol/water). Finally, the obtained particles were washed with EtOH (3 x 100 mL), dispersed in ethanol (50 mL) and stored in a refrigerator. For certain analysis prepared particles were dried under vacuum in an oven at 40 °C for 48 h. (R.R. Castillo, L. de la Torre, F. García-Ochoa, M. Ladero, M. Vallet-Regí, *International Journal of Molecular Science*, 2020, 21, 7899; T-W. Kim, P-W. Chung, V.S-Y. Lin, *Chemistry of Materials*, 2010, 22, 5093.)

Synthesis of nano-MCM-48.

0.5 g of CTAB and 2.05 g of F127 are dissolved in 96 mL of distilled water, 34 g of EtOH, and 10.05 g of 29 wt % ammonium hydroxide solution at room temperature. After complete dissolution, 1.8 g of TEOS is added into the mixture at once. After 1 min of mechanical stirring at 1000 rpm, the mixture was kept at a static condition for 24 h at RT for further silica condensation. The white solid product is recovered by ultrahigh speed centrifuge (Sorvall Evolution Centrifuge), washed with water, and dried at 343 K in air. The final template-free MCM-48 MSN materials are obtained after calcinations at 823 K in air. For the posthydrothermal treatment, the centrifuged sample is placed in the Teflon-lined autoclave with 8.5 mL of distilled water. The hydrothermal temperature varied from 403 to 423 K for 2 days in static conditions. (T-W. Kim, P-W. Chung, V.S-Y. Lin, *Chemistry of Materials*, 2010, 22, 5093.)