Supplementary Information

Interzeolite-Type Transformation between Microporous Titanosilicates

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Experimental Section

The synthesis of the parent ETS-4 crystals was carried out as follows: 4.43 g of colloidal silica (Ludox AS-40, Sigma-Aldrich) was dissolved in a solution containing 36 g of distilled water, 3.95 g of sodium hydroxide (NaOH, ≥98%, pellets, Sigma-Aldrich). To this solution, 1.1 mL of titanium tetrachloride ($TiCl_4$, Sigma-Aldrich), previously hydrolyzed in 20 g of distilled water, was added. The resulting white gel was homogenized for 30 minutes and then transferred to a Teflon-lined stainless-steel autoclave. The synthesis was carried out under static conditions in a preheated oven at 230 °C for 24 hours. After cooling the autoclave with running tap water, the product was filtered, washed several times with distilled water, and dried in air at 60 °C overnight. The transformation from ETS-4 to GTS-1 was performed in a Teflon-lined stainless steel autoclave. In this process, 0.48 g of as-synthesized ETS-4 was placed in a solution containing 1.93 g of potassium hydroxide (KOH, 90%, flakes, Sigma-Aldrich) and 20 g of distilled water (1.72 M solution). The mixture was then heated at 230 °C for a specified period. After heating, the product was recovered using the same procedure applied for the synthesis of ETS-4. The crystal structure and crystallinity of the samples were analyzed using powder X-ray diffraction (XRD) on a Bruker D8

diffractometer with Cu $K\alpha_{1,2}$ radiation. Powder XRD patterns were collected in θ–2θ mode over a 2θ range of 5–40°, with a step size of 0.05° and a counting time of 1 second per step. The crystallinity was calculated by DIFFRAC.EVA software comparing the integrated intensity of the XRD peaks of the parent and daughter phases. The framework density (FD) was calculated by the formula: FD=(number of framework atoms per unit cell)/(unite cell volume)x1000, where the unit cell volume is expressed in $Å^3$.

Sample morphology and chemical composition were examined using scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDS), performed on a NanoSEM-FEI Nova 200 system equipped with an EDAX Pegasus X4M detector.

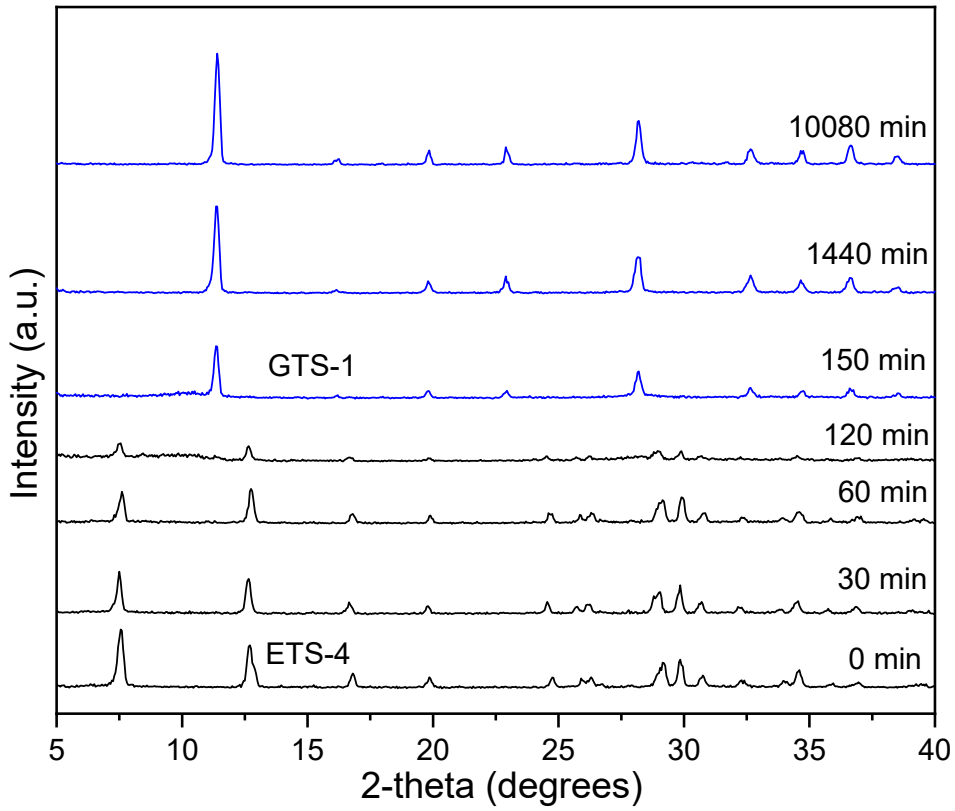


Figure S1. Powder XRD patterns showing ETS-4 to GTS-1 transformation followed up to 10080 min of synthesis time.

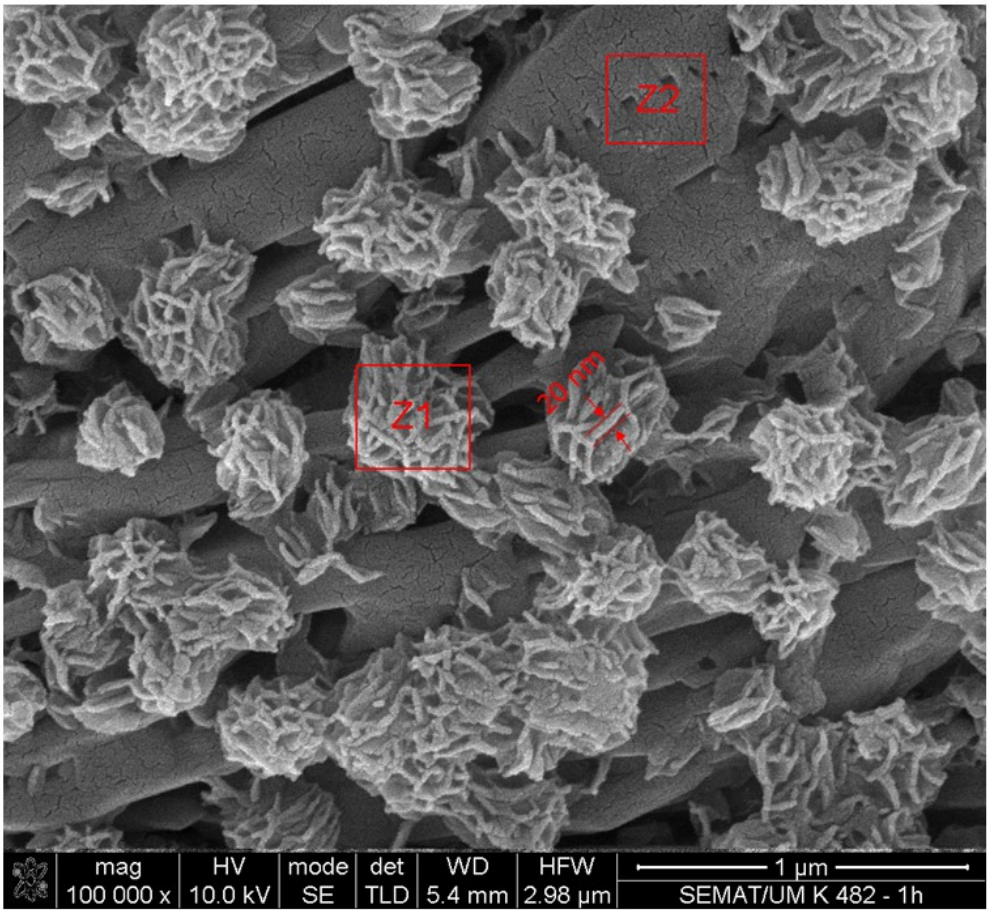
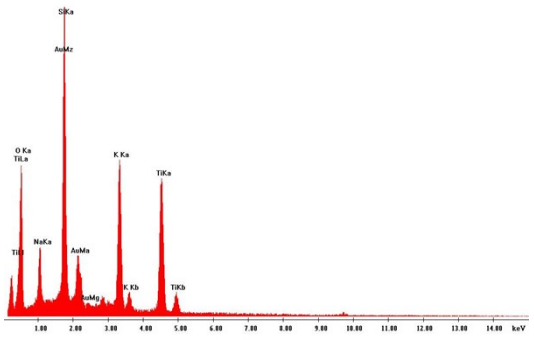


Figure S2. SEM image of ETS-4 treated for 60 min in KOH solution.

Elem	Wt %	At %	K-Ratio	Z	A	F
O K	34.90	54.72	0.0648	1.0706	0.1735	1.0002
NaK	4.86	5.30	0.0198	0.9986	0.4072	1.0017
SiK	17.88	15.97	0.1429	1.0202	0.7809	1.0031
K K	15.56	9.98	0.1459	0.9659	0.9535	1.0184
TiK	26.81	14.04	0.2324	0.8984	0.9650	1.0000
Total	100.00	100.00				

Elem	Wt %	At %	K-Ratio	Z	A	F
O K	33.87	53.72	0.0616	1.0729	0.1695	1.0002
NaK	4.98	5.49	0.0202	1.0007	0.4048	1.0016
SiK	17.51	15.82	0.1398	1.0226	0.7782	1.0032
K K	15.39	9.99	0.1448	0.9680	0.9537	1.0195
TiK	28.26	14.97	0.2457	0.9005	0.9653	1.0000
Total	100.00	100.00				



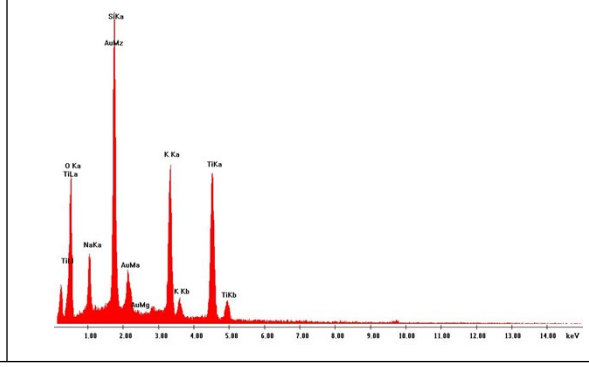


Figure S3. EDS analysis of layer-like nanoparticles (left; Z1) and uncovered surface (right; Z2) of ETS-4 after treatment for 60 min in KOH solution.

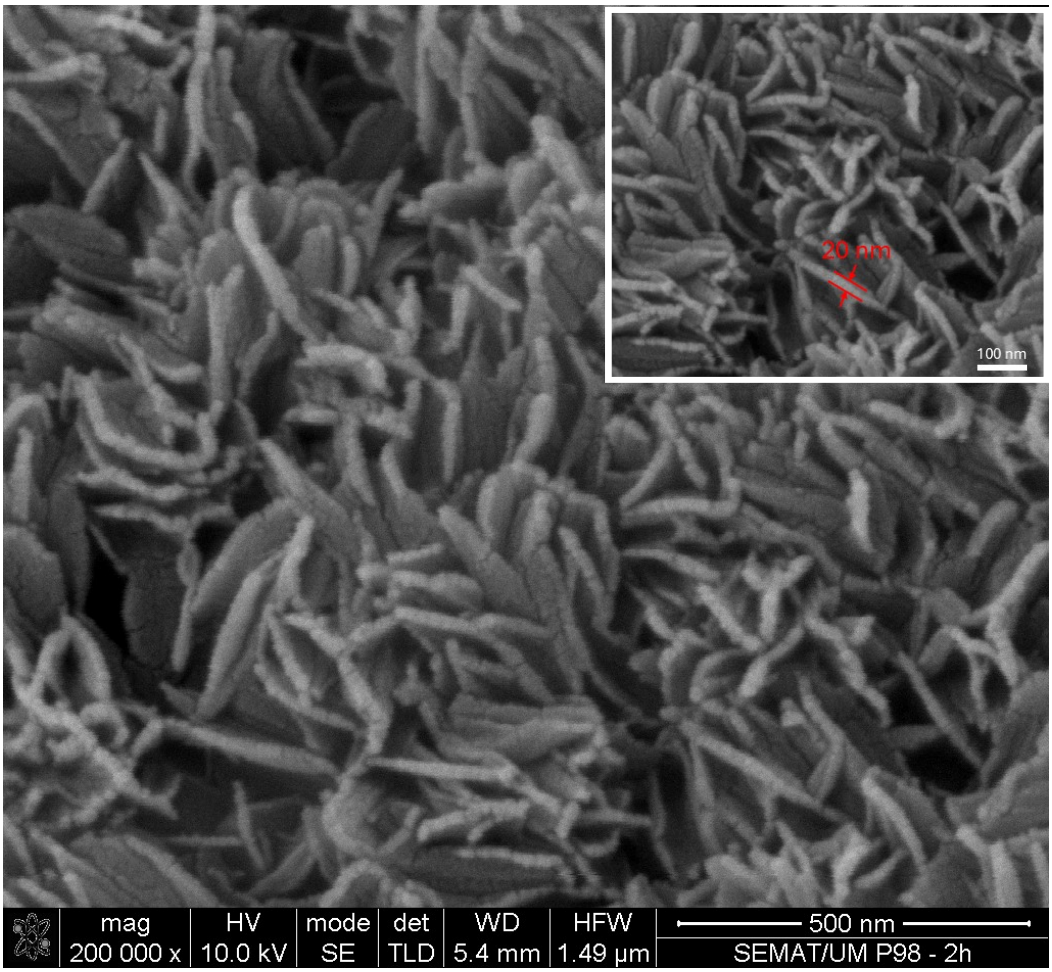


Figure S4. SEM image of ETS-4 after 120 min of treatment in KOH solution.

Elem	Wt %	At %	K-Ratio	Z	A	F
O K	26.79	44.22	0.0490	1.0741	0.1701	1.0002
NaK	4.54	5.21	0.0199	1.0018	0.4369	1.0026
SiK	28.31	26.62	0.2335	1.0241	0.8030	1.0028
K K	13.70	9.25	0.1262	0.9693	0.9352	1.0167
TiK	26.66	14.70	0.2310	0.9017	0.9609	1.0000
Total	100.00	100.00				

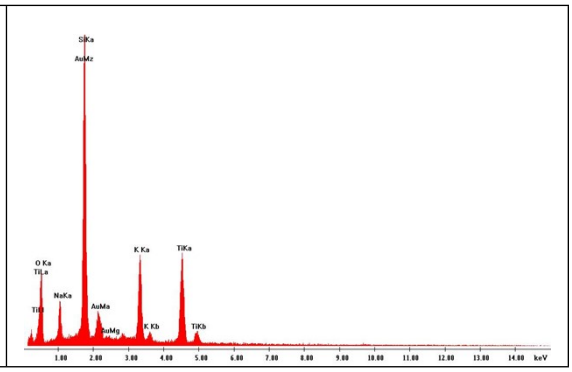
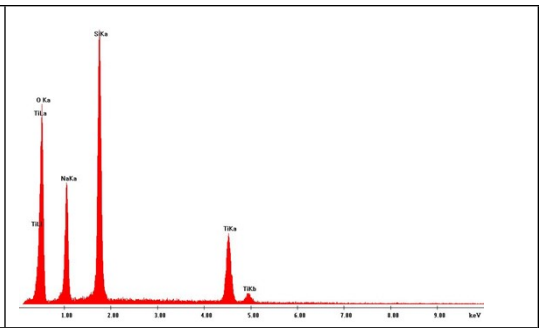


Figure S5. EDS analysis of ETS-4 after 120 min of treatment in KOH solution.

Elem	Wt %	At %	K-Ratio	Z	A	F
O K	43.95	60.10	0.1203	1.0498	0.2606	1.0004
NaK	13.10	12.47	0.0576	0.9794	0.4478	1.0019
SiK	24.29	18.92	0.1856	0.9973	0.7655	1.0009
TiK	18.65	8.52	0.1619	0.8794	0.9874	1.0000
Total	100.00	100.00				



Elem	Wt %	At %	K-Ratio	Z	A	F
O K	43.35	63.78	0.0898	1.0635	0.1948	1.0002
NaK	4.44	4.55	0.0179	0.9921	0.4049	1.0013
SiK	13.45	11.27	0.1067	1.0124	0.7816	1.0028
K K	12.22	7.36	0.1154	0.9592	0.9640	1.0207
TiK	26.52	13.03	0.2309	0.8920	0.9760	1.0000
Total	100.00	100.00				

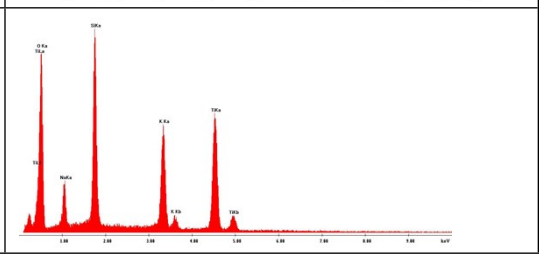


Figure S6. EDS analysis of the as-synthesized ETS-4 (up) and GTS-1 (down) obtained after 150 min of treatment in KOH solution (1.72 M).

Table S1. Experimental conditions of treatment of ETS-4 and the obtained run products.

No.	Time (min)	T °C	KOH (M)	Daughter phase
1	30	230	1.72	ETS-4
2	60	230	1.72	ETS-4
3	120	230	1.72	ETS-4
4	150	230	1.72	GTS-1
5	150	230	0.86	ETS-4
6	150	230	0.43	ETS-4
7	150	150	1.72	ETS-4
8	150	150	3.44	unknown
9	150	150	6.88	GTS-1
10	360	230	1.72	GTS-1
11	1440	RT	1.72	ETS-4
12	1440	230	1.72	GTS-1
13	10080	230	1.72	GTS-1

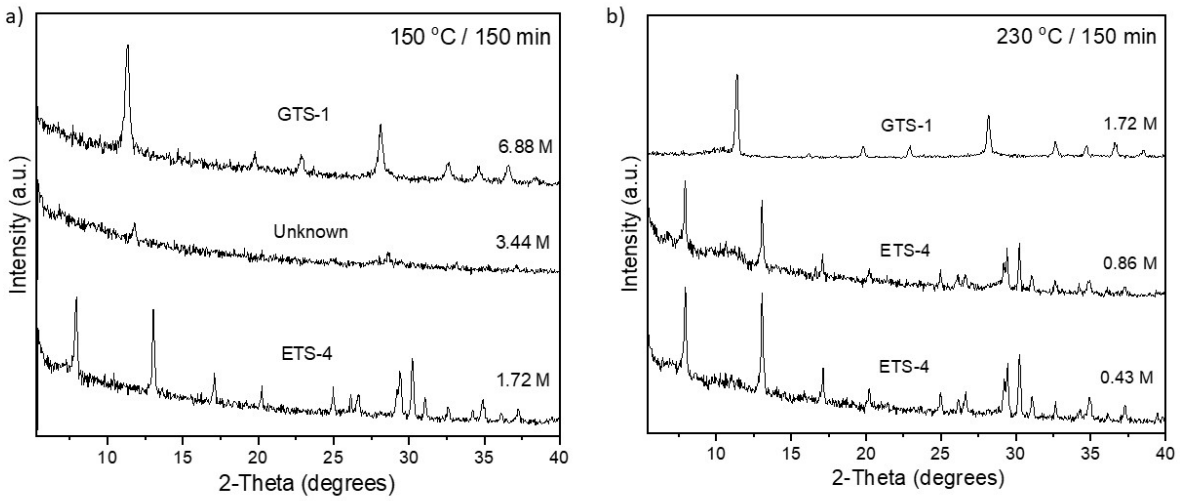


Figure S7. Powder XRD patterns showing the ETS-4 to GTS-1 transformation after 150 min at (a) 150 °C and (b) 230 °C under different KOH concentrations.

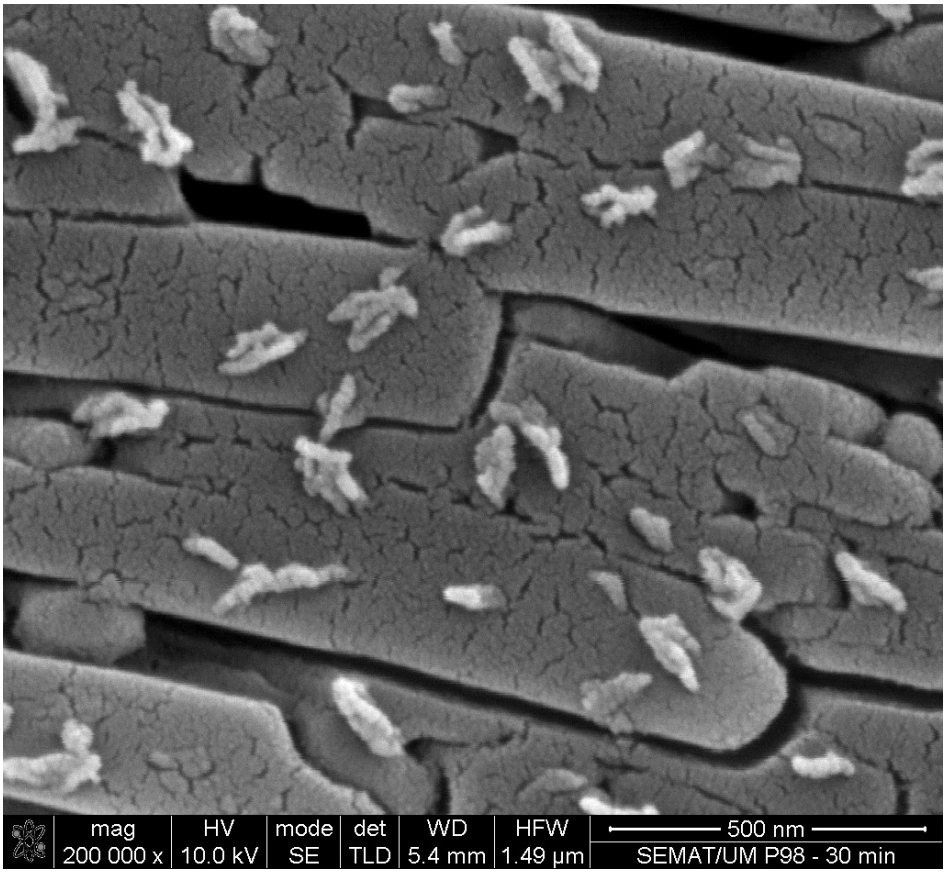


Figure S8. SEM image of ETS-4 treated at 230 °C for 30 min in KOH solution (1.72 M).

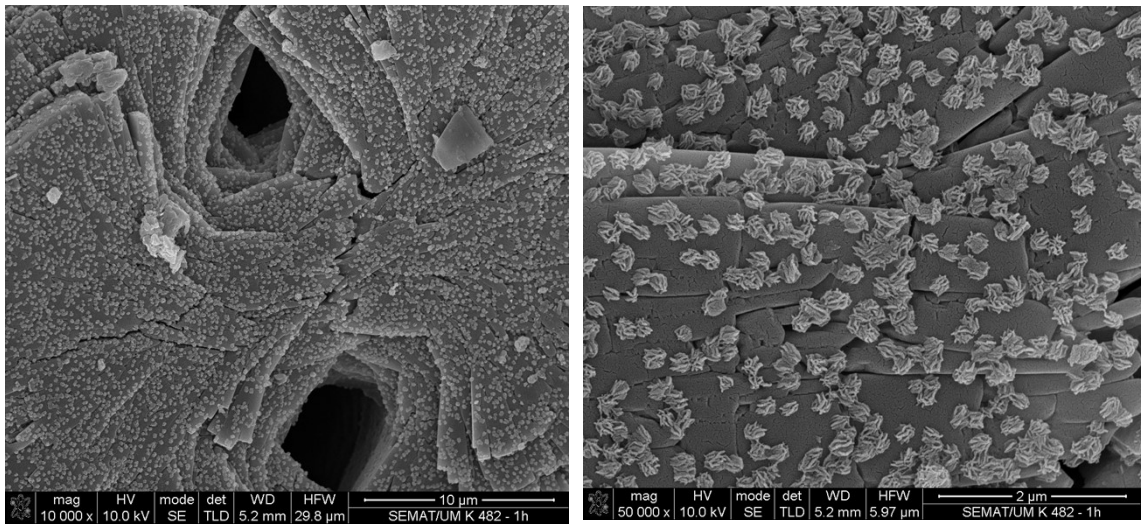


Figure S9. SEM images of ETS-4 treated at 230 ^{o}C for 60 min in KOH solution (1.72 M).

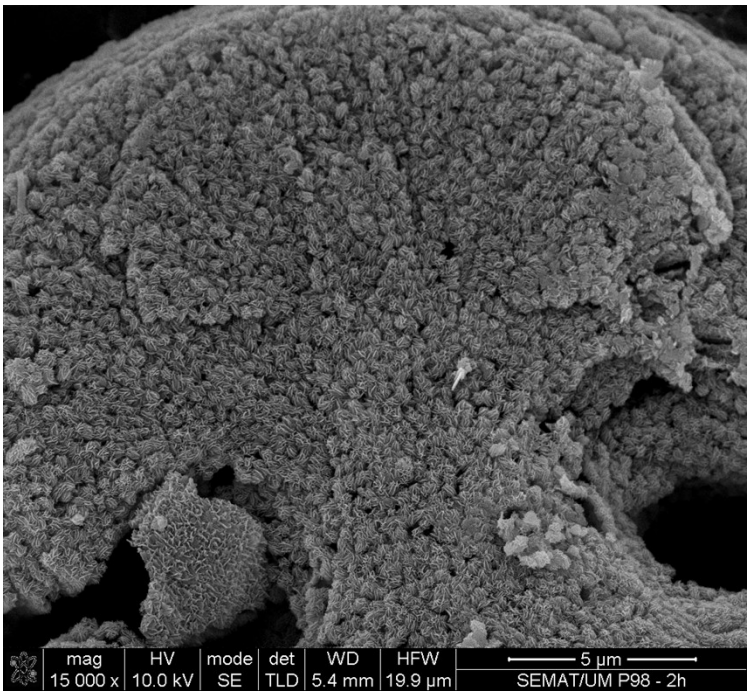


Figure S10. SEM image of ETS-4 treated at 230 ^{o}C for 120 min in KOH solution (1.72 M).