

Figure S1. DRIFT spectra recorded after adsorption of pyridine on MgSi-4 at 50°C with (top, in blue) and without pre-adsorbed water (bottom, in red).

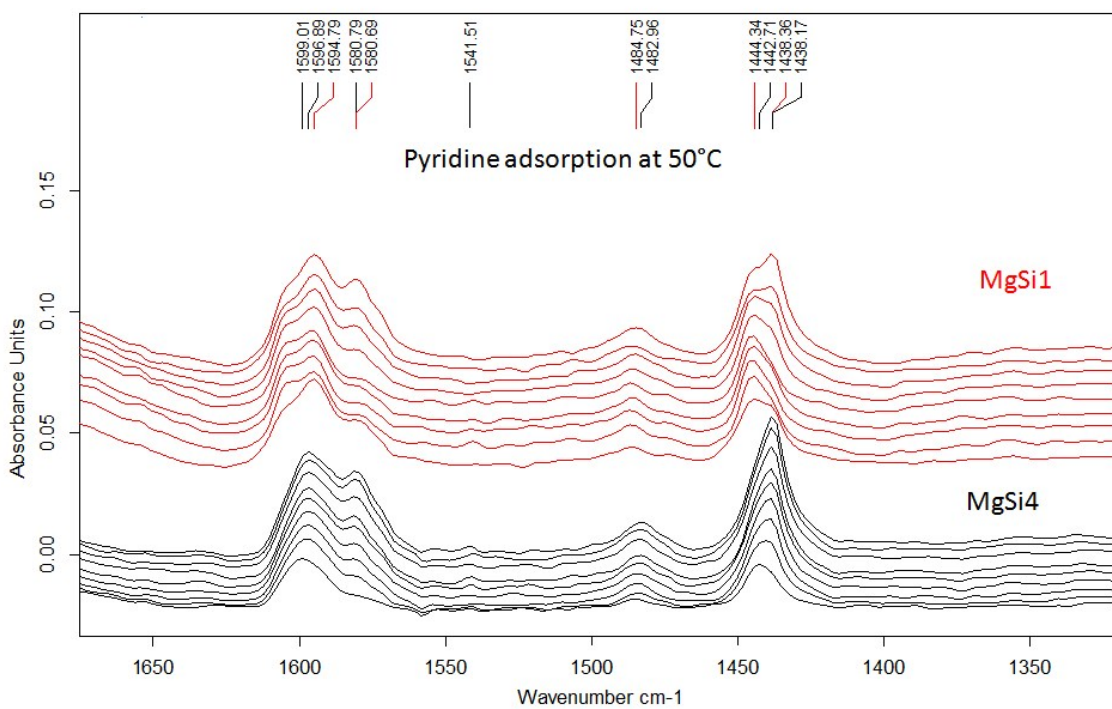


Figure S2. DRIFT spectra recorded in function of time during adsorption of pyridine at 50°C on MgSi-1 and MgSi-4.

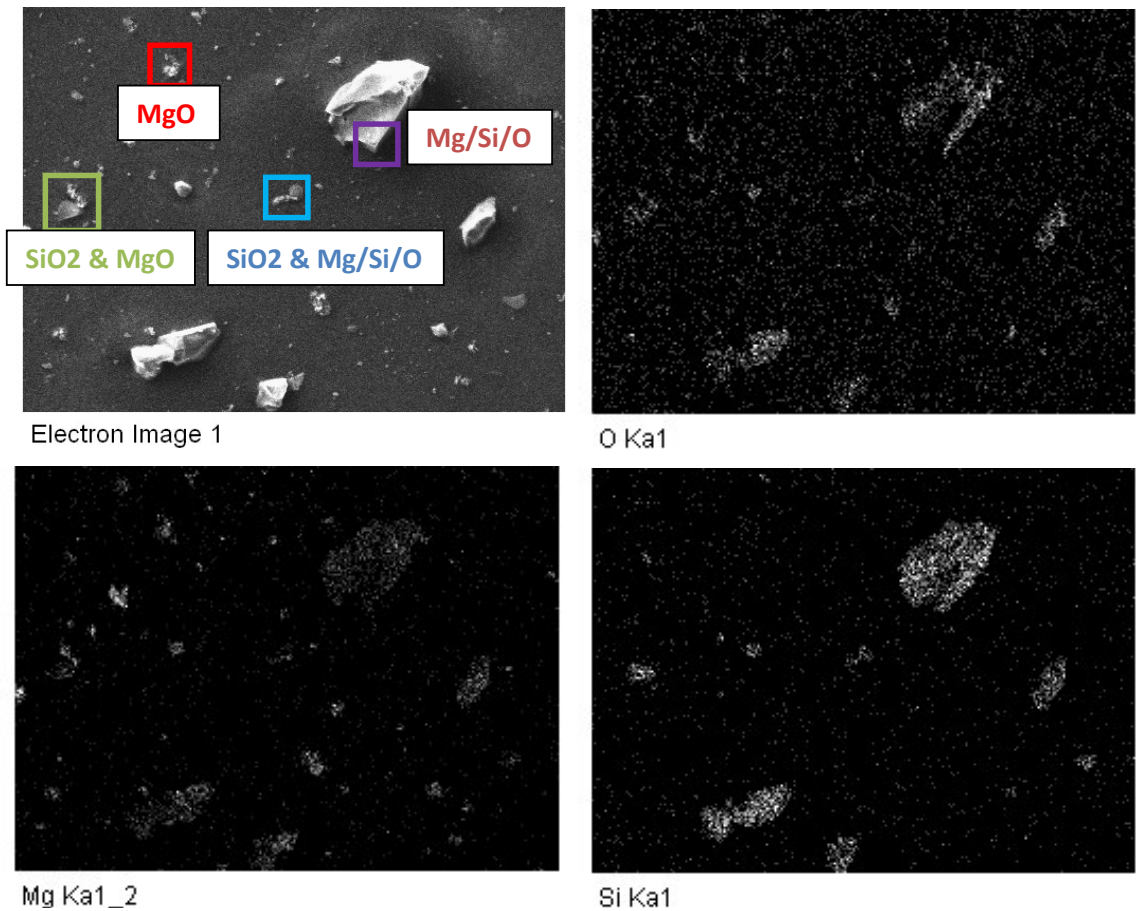


Figure S3. SEM-EDX analysis of MgSi-4.

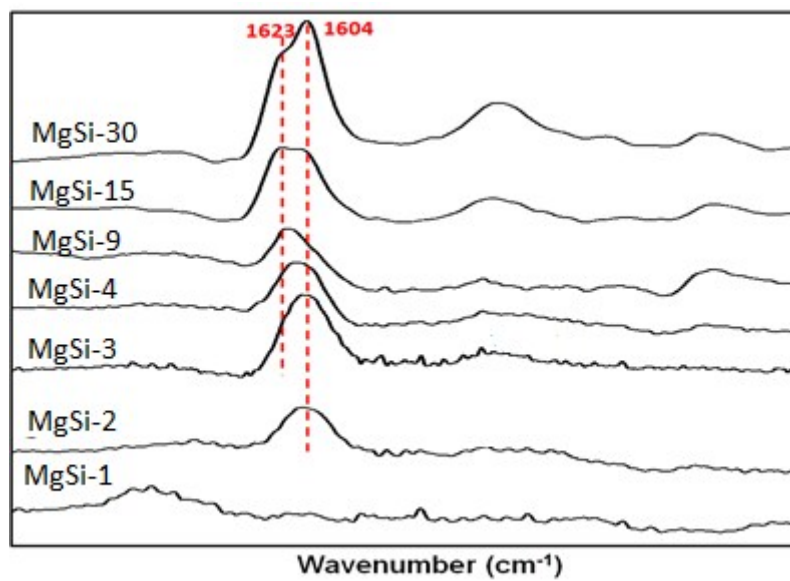


Figure S4. Zoom in the low wavenumber zone after ethanol feeding and thermal treatment at 400°C for the different MgSi materials

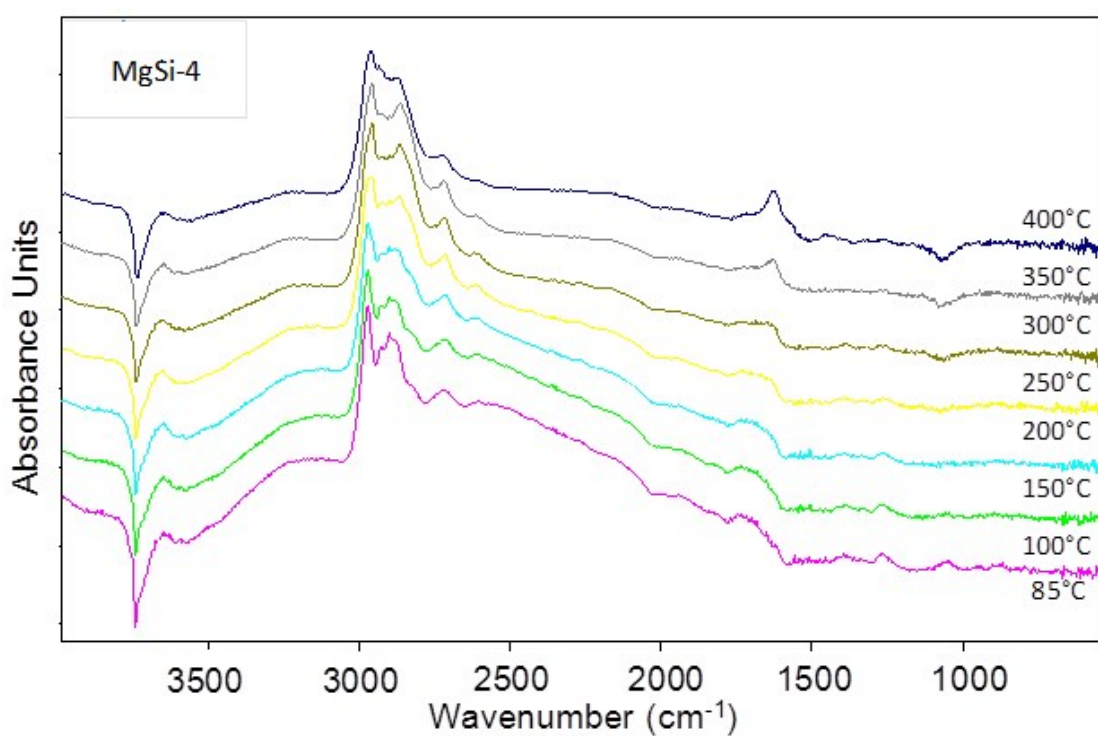
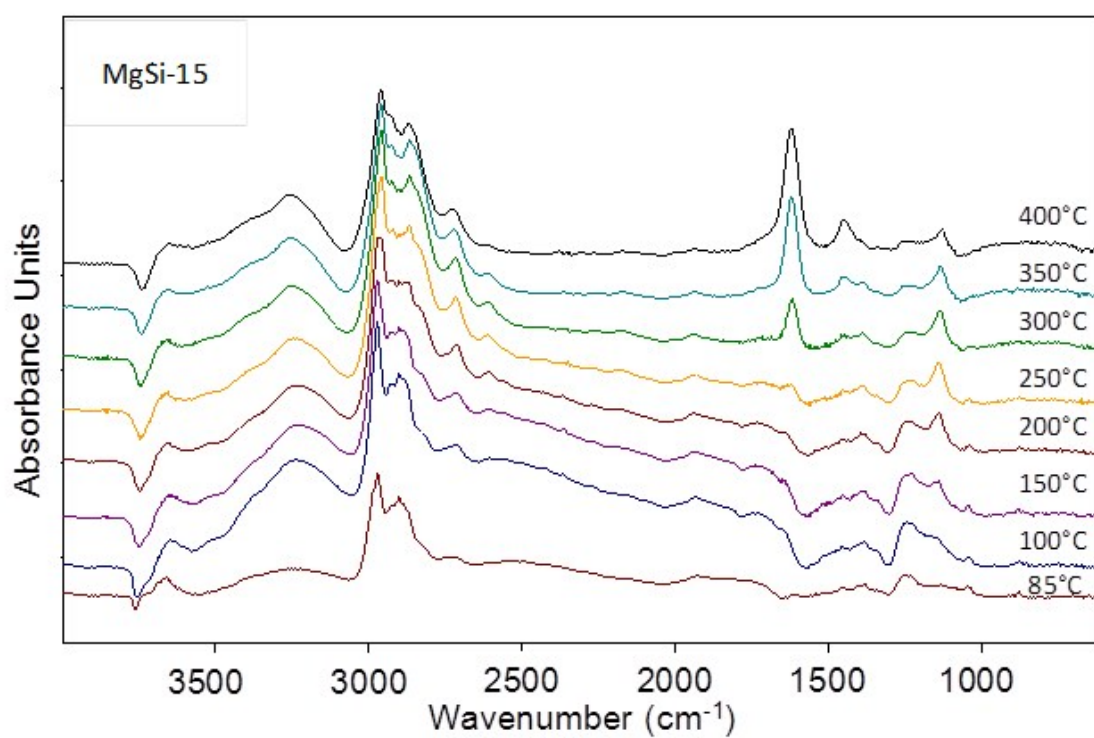


Figure S5. In-situ DRIFTS spectra of the MgSi-15 (Top) and MgSi-4 (Bottom) recorded while feeding ethanol and increasing temperature.

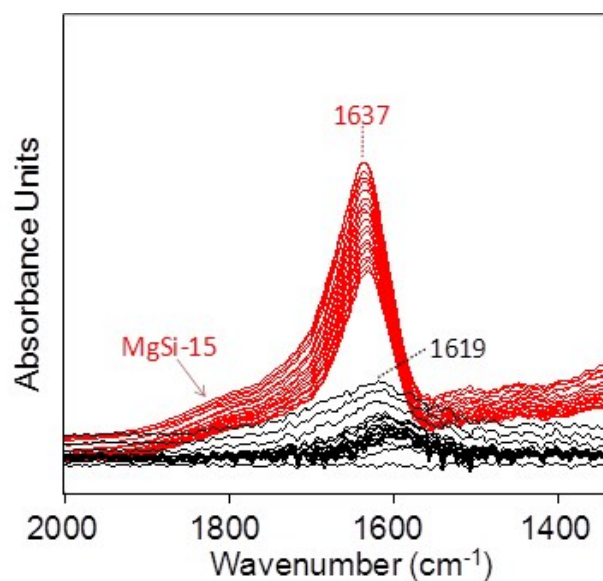


Figure S6. Adsorption of CO₂ at 400°C over MgSi-15 (Red) and MgSi-1 (Black).

Table S1. Results of catalytic experiments with catalysts at 400°C and 0.4 s contact time.

| Catalyst | SiO ₂ | MgSi-1 | MgSi-2 | MgSi-3 | MgSi-4 | MgSi-9 | MgSi-15 | MgSi-30 | MgO |
|------------------------|------------------|--------|--------|--------|--------|--------|---------|---------|------|
| Ethanol conv. (%) | 29.2 | 70.8 | 58.6 | 63.5 | 52.8 | 46.2 | 41.2 | 43.5 | 31.6 |
| Yield Butadiene (%) | 0 | 5.4 | 10.1 | 13.2 | 14.5 | 14.5 | 16.8 | 7.5 | 3.4 |
| Yield Ethylene (%) | 4.3 | 53.2 | 32.7 | 27 | 16.4 | 8.9 | 2.3 | 3.8 | 0.6 |
| Yield Butenes (%) | 0 | 0.7 | 2.8 | 4.0 | 2.8 | 1.4 | 0.5 | 0.1 | 0 |
| Yield propylene (%) | 0 | 0.2 | 1.4 | 2.4 | 1.5 | 0.8 | 0.3 | 0 | 0.1 |
| Yield Acetaldehyde (%) | 15.4 | 2.8 | 2.4 | 3.0 | 2.3 | 2.2 | 2.4 | 2.0 | 3.6 |
| Yield Acetone (%) | 0.5 | 1.1 | 1.9 | 3.0 | 1.9 | 3.7 | 3.1 | 1.9 | 3.6 |
| Yield Diethylether (%) | 5.3 | 7 | 4.6 | 4.1 | 2.9 | 1.6 | 1.1 | 0.7 | 0.4 |
| Yield Butanal (%) | 0 | 0 | 0 | 0 | 0.1 | 0 | 1.3 | 0.2 | 0 |
| Yield Ethylacetate (%) | 0.3 | 0 | 0 | 0 | 0.1 | 0.9 | 1.5 | 0.4 | 0.5 |
| Yield Alkenols (%) | 0 | 0 | 0.1 | 0.4 | 0.5 | 1.7 | 1.3 | 2.4 | 0 |
| Yield 1-butanol (%) | 0 | 0 | 0.1 | 0.2 | 0.4 | 3.6 | 5.3 | 2.2 | 10.5 |
| Mass balance (%) | 88 | 99 | 96 | 90 | 82 | 85 | 87 | 49 | 72 |