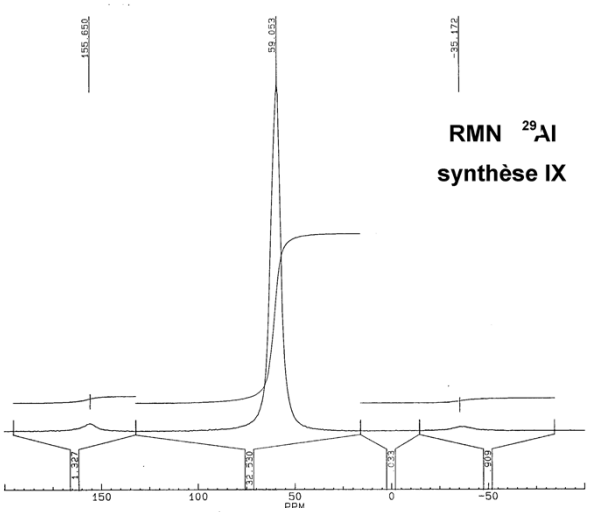
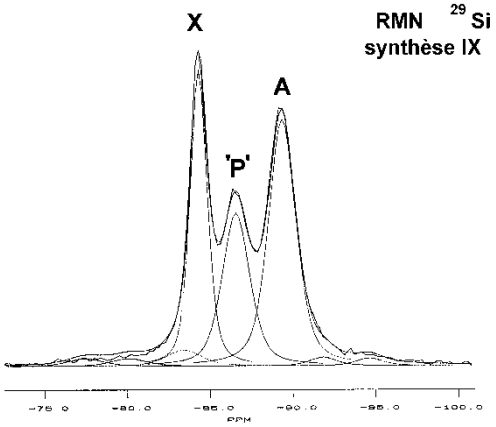
SUPPLEMENTARY MATERIALS



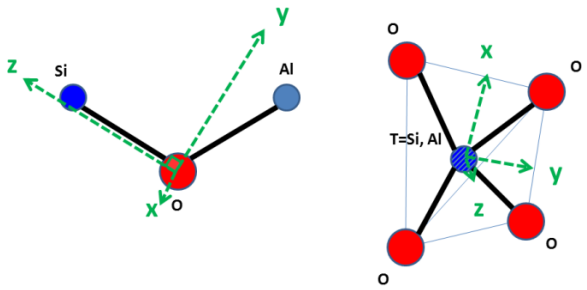
Supplementary Fig. 1b ^{27}Al NMR spectrum on dehydrated powder showing only the peak at 70ppm characteristic of Al in tetrahedral coordination. The absence of the signal at 4 ppm related to the presence of octahedral Al and hydrolysis of the framework testify of the preservation



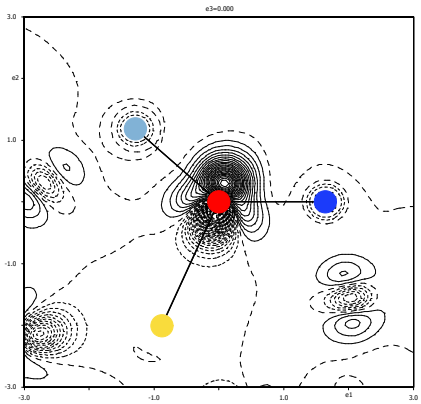
Supplementary Fig. 1b ^{29}Si NMR spectrum on the raw synthesized powder. Only the Q4(4Al) peaks of the three phases (A, X and P zeolites) forming the powder are present. The peak areas reflect the mixing ratio of the phases.

Supplementary Table 1 n and ζ (Bohr^{-1}) values used in multipolar refinement

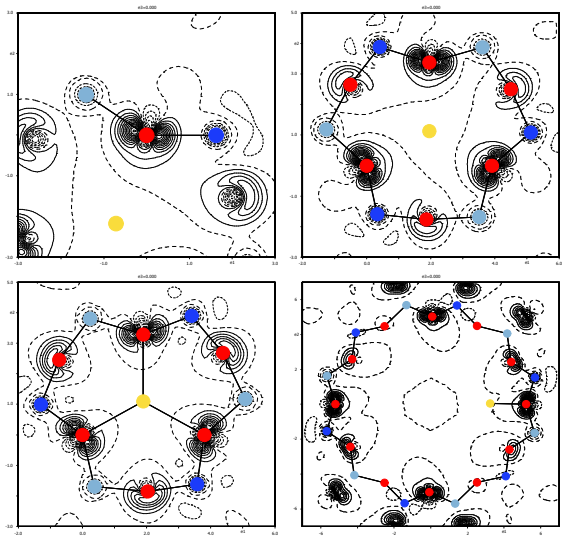
Plm order	1	2	3	4	5	6	7	8
n(Si)	4	4	4	6	8	10	12	14
ζ(Si)	3.063	3.063	3.063	3.063	3.063	3.063	3.063	3.063
n(Al)	4	4	4	6	8	10	12	14
ζ(Al)	2.733	2.733	2.733	2.733	2.733	2.733	2.733	2.733
n(O)	2	2	2	3	4	5	6	7
ζ(O)	4.466	4.466	4.466	4.466	4.466	4.466	4.466	4.466



Supplementary Fig. 2 Local frames for multipolar parameters



Supplementary Fig. 3 Static deformation density at Step 3c, after releasing the equivalence constraint on oxygen Plm parameters. CRM^2 dataset, contours of 0.1 e/Å^3.



Supplementary Fig. 4 Static deformation density maps calculated at Step 3b (ESRF dataset) in the Si-O2-Al plane, the O2-O2-O2 plane of the hexagonal prism, the O3-O3-O3 plane of the hexagonal window and the average plane of the dodecagonal window. The Si, Al, O and Na atoms are symbolized by blue, light blue, red and yellow disks. Only one of the Na3 cations of the dodecagonal window is shown. contours of 0.1 e/Å^3.