EXPERIMENT MA4489

1. Total scattering experiment: samples collected at room temperature

Total scattering intensities were recorded by a Perkin Elmer 2D detector. The capillary was translated 15 times, for each point 90 images were collected with an exposure time of 0.5 sec.

2. Results: qualitative comparison

Pair distribution functions were obtained, for a first evaluation, by PDFgetX3 software using *Qmax*=21.

2.1 EZ-zeolites *vs* amorphous

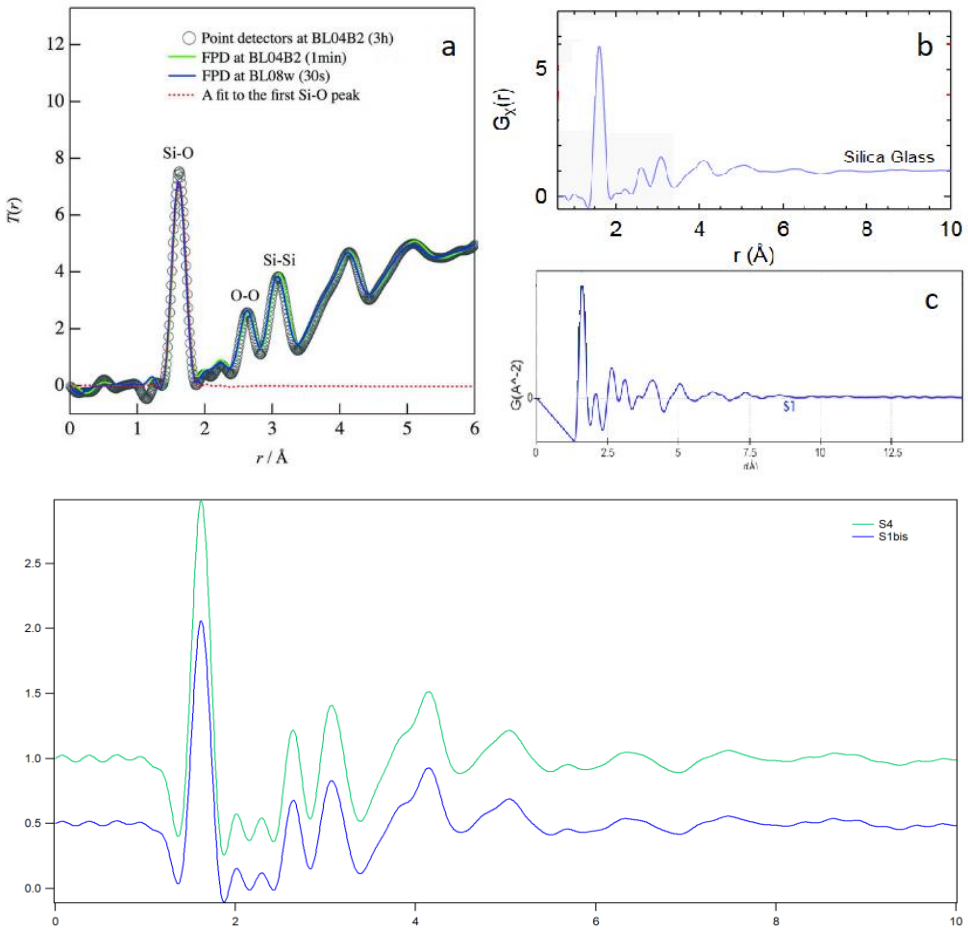

Figure 1. Top panel: qualitative comparison of samples EZ-CHA-2 named S1bis (blue line) and EZ-MFI-2 named S4 (green line). Radius (Å) of the PDF is reported on the X axis, while the Y axis is expressed in an arbitrary unit. Bottom panel: Pair Distribution Function of amorphous silica from a) Ohara et al., 2018, Journal of Synchrotron Radiation, 25(6), 1627 b) Khouchaf et al., 2020, Materials 13(19), 4393 c) "The Study of Disorder in Amorphous Silica, Alkali-Silica Reaction Gel and Fly Ash", thesis of Çağla Meral, University of California, Berkeley, 2012.

As expected, the local structure from 1 to ~4.3 Å is very similar for embryonic samples and the amorphous (Figure 1). However, the peak at around 5 Å seems better resolved and intense in the embryonic zeolites. This Si-O distance occurs between at least three tetrahedra, thus it is possible that

in embryonic zeolites the probability of finding this distance is higher, may mining more tetrahedra involved (Figure 2). In addition, from Figure 1, we can observe a different dampening of the signal, with a slightly faster decrease for the amorphous. This is a good indication that embryonic zeolites have a longer coherent correlation length, (*i.e.* longer short range order).

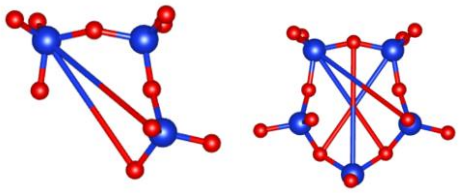

Figure 2. On the left panel, distance Si-O =5.03 Å is highlighted, this distance can exist if at least three tetrahedra are positioned as reported, but more tetrahedra can be involved even, in theory, forming a pentagon (right panel).

Considerations reported point out that the local structure of embryonic zeolites can be described with a peculiar atomic arrangement.

3.3 EZ-MFI samples doped with different samples

Figure 3 shows a qualitative comparison of EZ-MFI samples with similar Si/Al ratio, but with different doping. The first sample is not doped, while the last four present the 5 wt % of MnO_3, CuO, Fe_3O_4 and ZnO, respectively.

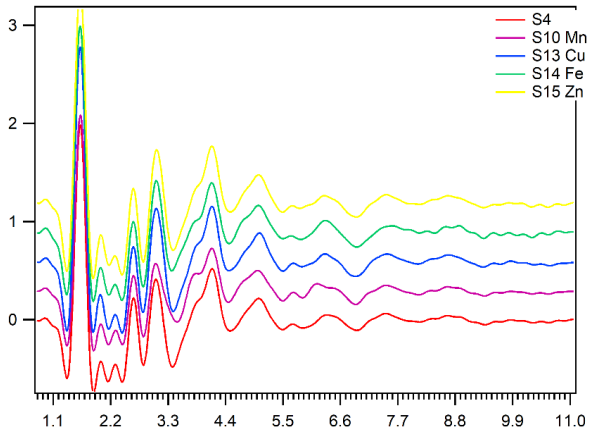

Figure 3. Qualitative comparison of samples EZ-MFI samples with similar Si/Al ratio, but with different. Radius (Å) of the PDF is reported on the X axis, while the Y axis is expressed in arbitrary unit.

Unfortunately, for all samples, the first distance metal-oxygen, which is around 1.9-2 Å, is hidden by the noise (i.e. termination ripples). The local structure appears similar for all samples up to around 5.5 Å, indeed, despite some variations in the intensity, the positions of the peaks are almost the same. Above 5.5 Å, instead, small differences can be observed.

Future analysis will be performed by EPSR software to give quantitative results on the local structure.