Identifying low and high density amorphous phases during zeolite amorphisation using small and wide angle x-ray scattering

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In situ experiments following the thermal amorphisation of zeolites reveal massive increases in small angle x-ray scattering (SAXS), persisting well beyond the stage where wide angle X-ray scattering (WAXS) can detect that any crystalline phase is present [1]. This heterogeneity in the amorphised phase can be attributed to the transition from a low density amorphous phase (LDA) to a high density amorphous phase (HDA) at the glass transition. The fractions of zeolite, LDA and HDA phases have been obtained from SAXS analysis using the development of the integrated intensity with progressive amorphisation and the corresponding changes in density – see Figure. These proportions of crystalline and low and high density amorphous phases can be used to interpret non-linear changes detected in the fractions of ^{n}Q species evident in ^{29}Si solid state NMR during zeolite amorphisation. Whilst the HDA phase is chemically disordered, the LDA phase exhibits much of the Al-Si ordering present in the starting zeolite. These findings are consistent with the low entropy perfect glasses predicted by Kauzmann to occur when super strong liquids are supercooled.

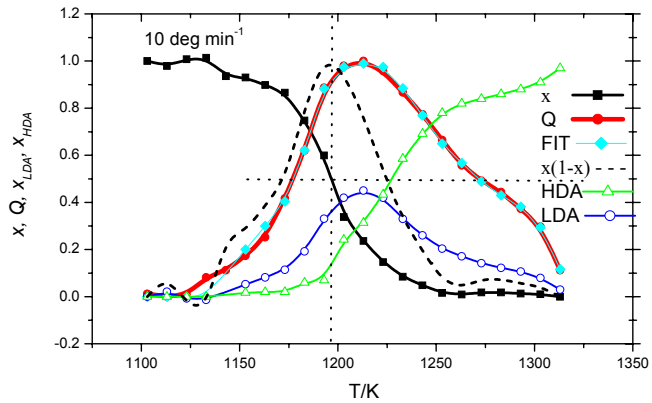

Figure. Amorphisation of Na zeolite Y followed by WAXS and SAXS, showing the proportions of low and high density amorphous phases, assuming liquid-liquid coexistence.

[1] Identifying low and high density amorphous phases during zeolite amorphisation using small and wide angle x-ray scattering. Meneau F and Greaves GN, *Nucl. Instrum. Meth. B* in press 2005