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Supplementary Information

Monitoring the Assembly-Disassembly-Organisation-Reassembly Process of Germanosilicate UTL through *in situ* Pair Distribution Function Analysis

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1. Initial total scattering intensity, I(Q) data for Ge-**UTL** after:
    a. 8 hr hydrolysing in water
    b. 15 hr hydrolysing in 6 M hydrochloric acid
    c. 12 hr hydrolysing in 12 M hydrochloric acid
2. The total scattering structure function, S(Q) data for Ge-**UTL** after:
    a. 8 hr hydrolysing in water
    b. 15 hr hydrolysing in 6 M hydrochloric acid
    c. 12 hr hydrolysing in 12 M hydrochloric acid
3. Reduced Pair Distribution Function, G(*r*) data out to high r-range (30 Å) for Ge-**UTL** after:
    a. 8 hr hydrolysing in water
    b. 15 hr hydrolysing in 6 M hydrochloric acid
    c. 12 hr hydrolysing in 12 M hydrochloric acid
4. Structural differences between calculated and experimental IPC-1P, through PDF refinement

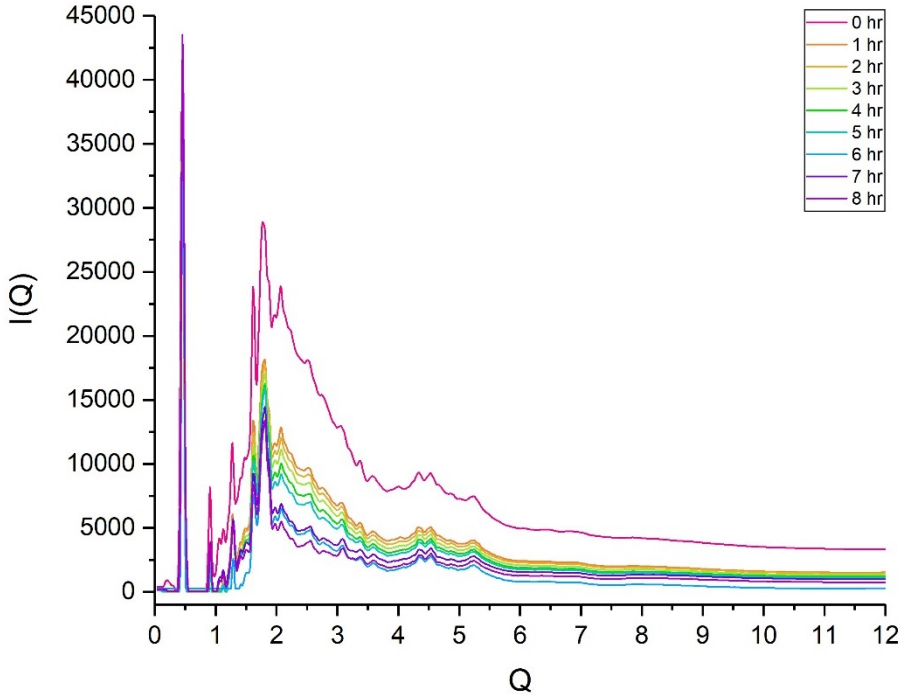


*Figure 1.a. Initial I(Q) data for Ge-**UTL** in water after hydrolysing for 8 hr at 100 ˚C. Data cut to Q = 12 to show the tail off of realistic data and key features <7. Where $I(Q) = I_{coh}(q) + I_{incoh}(q)$.*

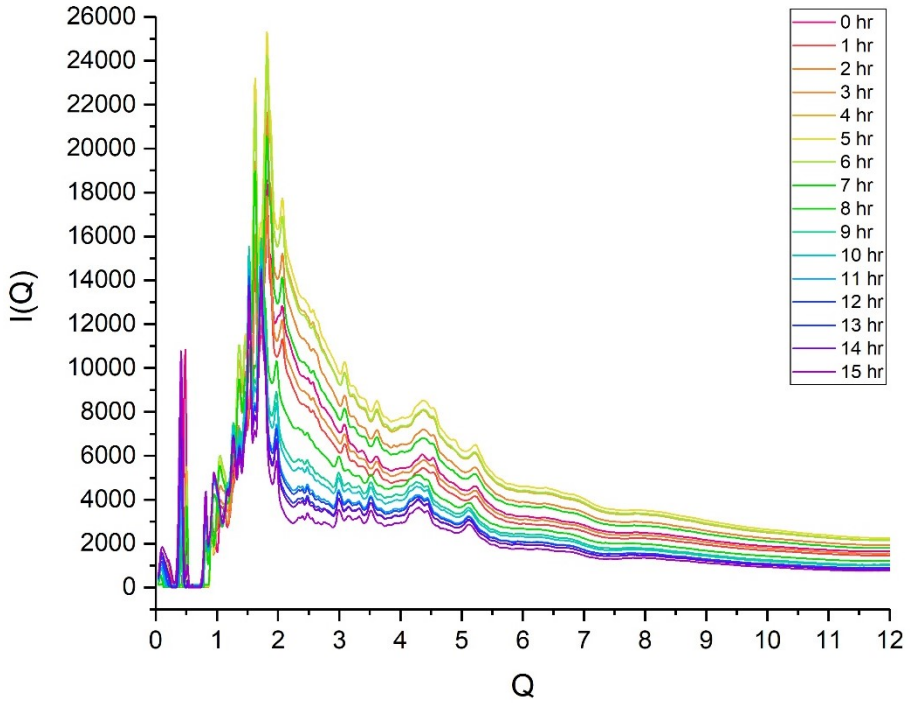


*Figure 1.b. Initial I(Q) data for Ge-**UTL** in 6 M hydrochloric acid after hydrolysing for 15 hr at 100 ˚C.*

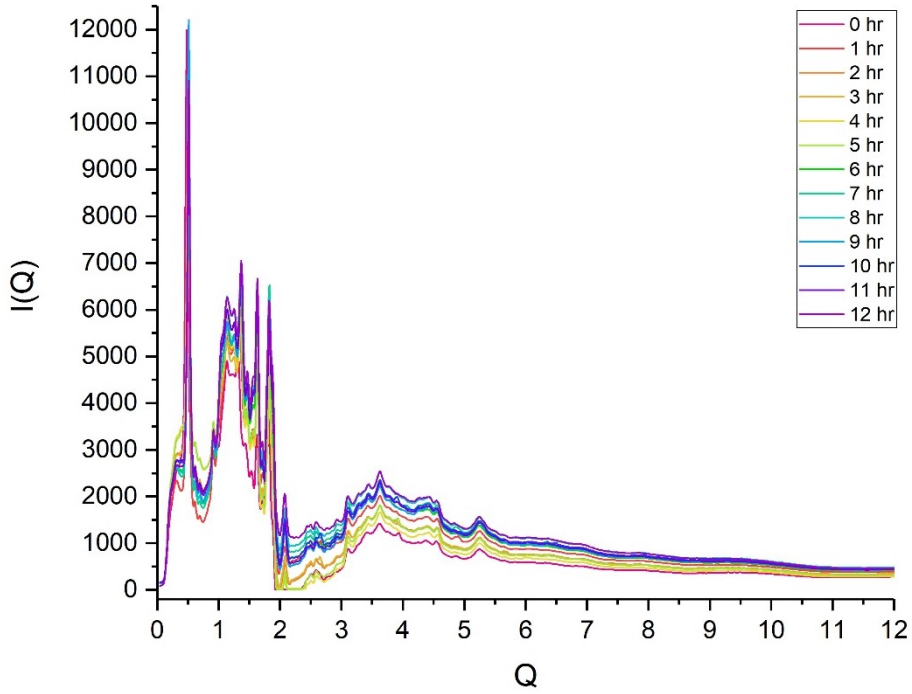


*Figure 1.c. Initial I(Q) data for Ge-**UTL** in 12 M hydrochloric acid after hydrolysing for 12 hr at 50 ˚C.*

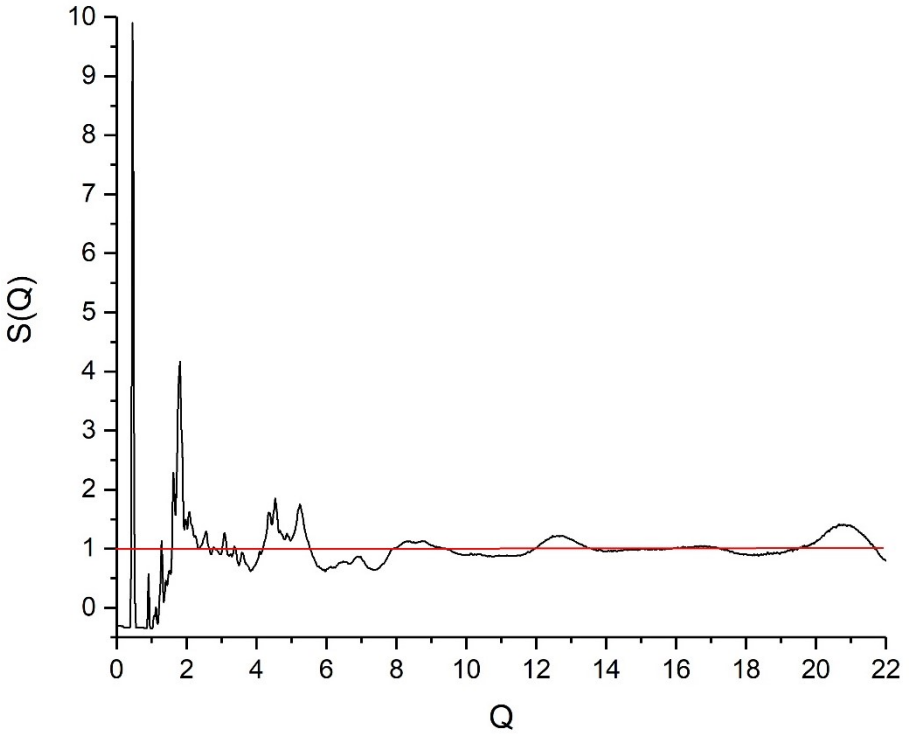
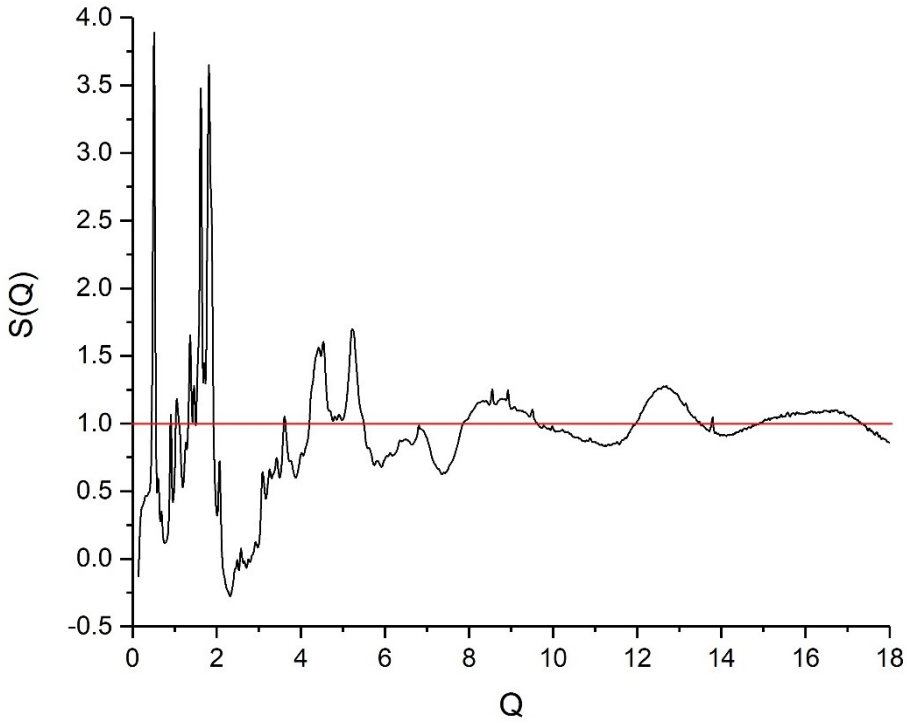


*Figure 2.a. S(Q) data for Ge-**UTL** in water after hydrolysing for 8 hr at 100 ˚C. S(Q) data oscillates around 1.*



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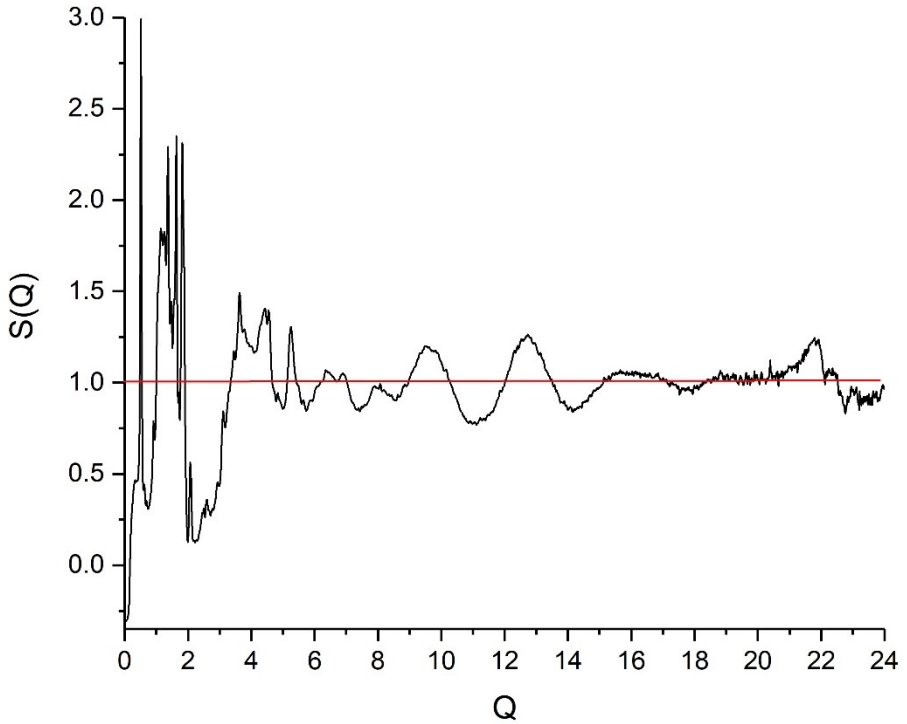

*Figure 2.c. S(Q) data for Ge-**UTL** in 12 M hydrochloric acid after hydrolysing for 12 hr at 50 ˚C. S(Q) data oscillates around 1.*

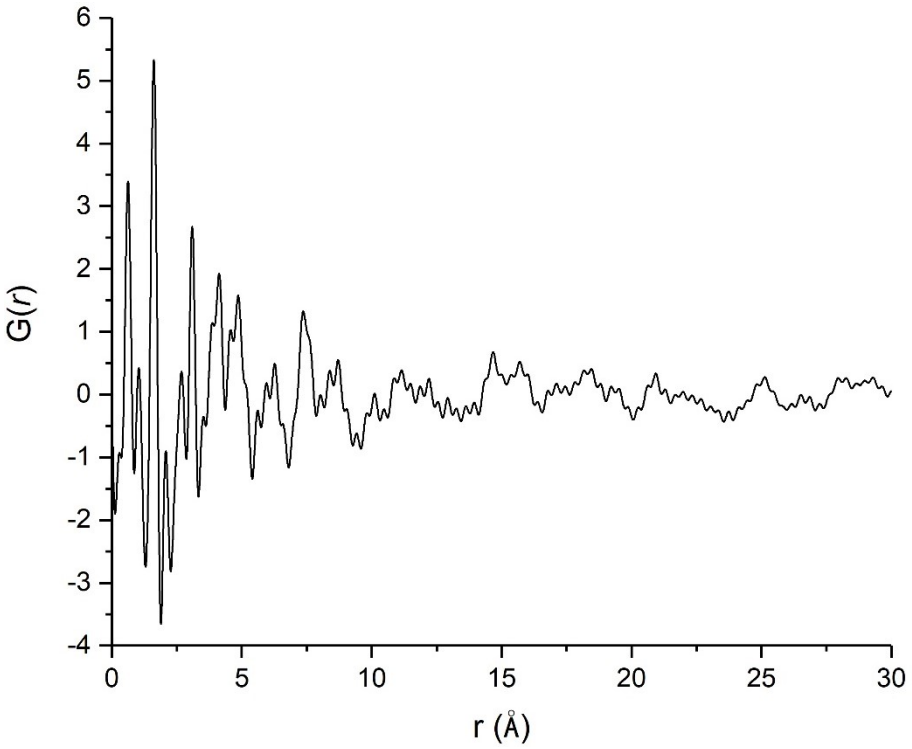

*Figure 3.a. Pair distribution function, G(r) data for Ge-**UTL** after hydrolysing for 8 hr in water at 100 ˚C. No peaks are observed after 12 Å, due to a lack of long-range order.*

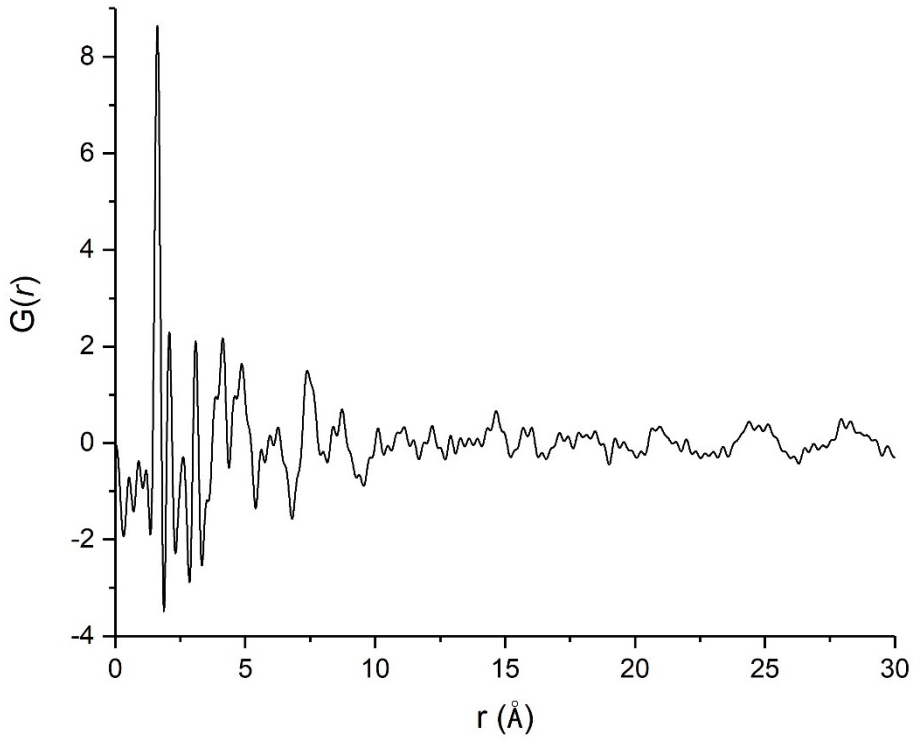

*Figure 3.b. Pair distribution function, G(r) data for Ge-**UTL** after hydrolysing for 15 hr in 6 M hydrochloric acid at 100 ˚C. No peaks are observed after 10 Å, due to a lack of long-range order. G(r) has minimal features at unphysical distances (r < 1 Å).*

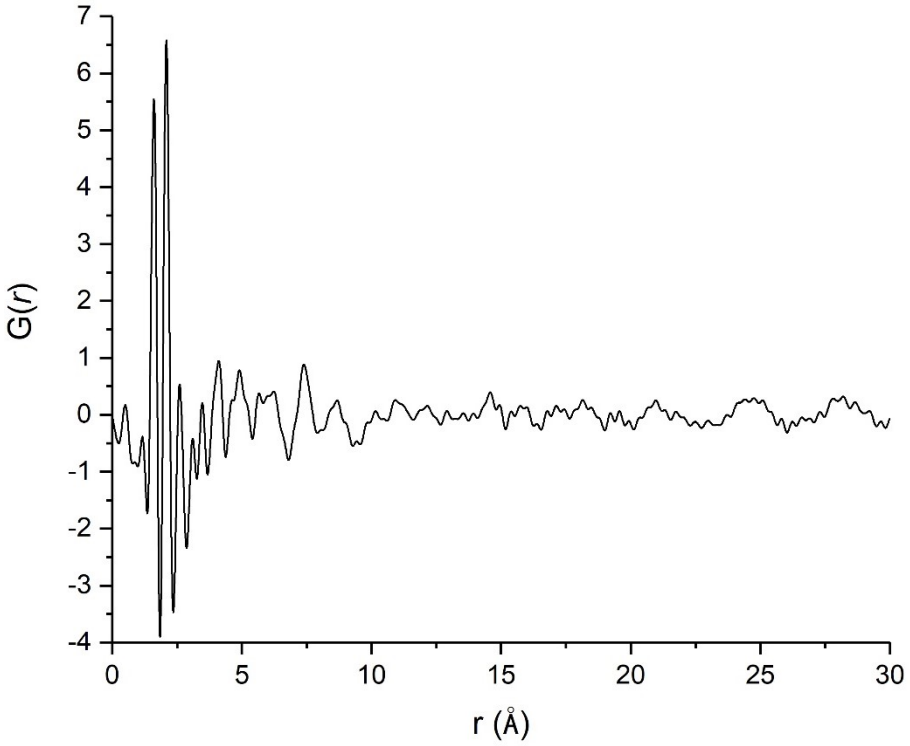

*Figure 3.c. Pair distribution function, G(r) data for Ge-**UTL** after hydrolysing for 12 hr in 12 M hydrochloric acid at 50 ˚C. No peaks are observed after 12 Å, due to a lack of long-range order. G(r) has minimal features at unphysical distances (r < 1 Å).*

Table 1. A structural comparison of calculated and experimental IPC-1P, through PDF refinement.

	Calculated IPC-1P	***Experimental IPC-1P***
Average Si-O bond lengths / Å	*1.61*	*1.63*
a / Å	*14.32*	*14.45*
b / Å	*13.90*	*14.12*
c / Å	*12.12*	*12.50*
α / ˚	*90.49*	*90.03*
β / ˚	*115.22*	*117.06*
γ / ˚	*120.23*	*119.89*