Appendix 1.

Diffusion coefficients for sodium carbonate (NC) have been calculated from the classic molecular dynamics simulations of Wilson et al. 2018. These simulations use the flexible anion approach applied to high energy X-ray diffraction data (Wilding et al 2016) augmented by fluctuation of charge. The carbonate anion flexibility and charge distribution can be varied. The key parameters that control the structure and dynamics of these carbonate liquids can be explored and compared to the liquid structures derived from X-ray experiment. The diffusivities of sodium and carbonate ions are calculated and so the fragility, structure and charge distribution of the simulated liquids can be explored. These simulations indicate that as charge separation increases the liquids there is formation of low-dimensional structures the extent of which are temperature-dependent and result in an increase in liquid fragility.

The diffusion data for different degrees of charge separation, $\Delta q = q_C - q_O$ ranging from ~-07e (q_C = -0.33, q_O = -1) to ~2.5e (q_C = -1.22, q_O = -1.36) are shown in table A1. These data are presented in Wilson et al 2018 in figure 3a. The viscosity is calculated from the Stoke's-Einstein equation.

$$\eta = \frac{k_B \mathrm{T}}{2\pi D d} \quad \text{(A1)}$$

With η viscosity in Pa*s, D the diffusion coefficient in m^2/s, d the mean interatomic separation obtained from the partial radial distribution functions (established as 3.5 Å, the average distance between Na and C obtained from the partial radial distribution function). Temperature is T and k_B is Boltzmann's constant. The diffusion of sodium and carbon atoms is correlated in these liquids so the diffusion and hence viscosity is calculated as an average of the two diffusion coefficients.

The structures of the liquid matches with a charge separation of charge (Δq = 2.28) and this fragile liquid shows a temperature-dependence of formation of extensive carbonate structures, the VFT curves for all the NC liquids are shown compared with that of the K_2CO_3-$MgCO_3$ liquid that is the focus of this study and experimental Na_2CO_3 liquid data in figure A1, the fraction of carbonate $(CO_3^{2-})_3$ triangles as a function of temperature and charge separation is also shown (Wilson et al., 2018).

Table A1: Sodium carbonate diffusion coefficients and calculated viscosity for different values of charge separation (Wilson et al. 2018)

Δq(e) -0.68

	Sodium			Carbon			Average	
Temperature (K)	Log_{10} D(cm^2/s)	Log_{10} η (Pa.s)	Error	Log_{10} D (cm^2/s)	Log_{10} η(Pa.s)	Error	Log_{10} η (Pa.s)	Error
800	-5.441	-1.888	-0.490	-6.303	-1.026	-0.567	-1.457	-0.528
900	-5.059	-2.219	-0.455	-5.939	-1.338	-0.535	-1.779	-0.495
1000	-4.802	-2.430	-0.336	-5.765	-1.467	-0.404	-1.948	-0.370
1100	-4.609	-2.581	-0.230	-5.414	-1.777	-0.271	-2.179	-0.251
1200	-4.407	-2.746	-0.176	-5.219	-1.934	-0.209	-2.340	-0.193
1300	-4.260	-2.858	-0.128	-5.000	-2.118	-0.150	-2.488	-0.139
1400	-4.203	-2.883	-0.126	-4.984	-2.102	-0.150	-2.493	-0.138
1500	-4.090	-2.966	-0.123	-4.709	-2.347	-0.141	-2.657	-0.132
1600	-4.022	-3.006	-0.080	-4.582	-2.446	-0.092	-2.726	-0.086
1750	-3.907	-3.082	-0.039	-4.507	-2.482	-0.045	-2.782	-0.042
2000	-3.727	-3.205	-0.037	-4.223	-2.708	-0.042	-2.956	-0.040
2500	-3.582	-3.252	-0.036	-3.937	-2.897	-0.039	-3.075	-0.038

Δq(e) 0.2

	Sodium			Carbon			Average	
Temperature (K)	Log_{10} D(cm^2/s)	Log_{10} η (Pa.s)	Error	Log_{10} D (cm^2/s)	Log_{10} η(Pa.s)	Error	Log_{10} η (Pa.s)	Error
800	-5.447	-1.882	-0.490	-6.016	-1.313	-0.541	-1.597	-0.516
900	-5.150	-2.128	-0.464	-5.823	-1.455	-0.524	-1.791	-0.494
1000	-4.822	-2.411	-0.338	-5.599	-1.633	-0.392	-2.022	-0.365
1100	-4.675	-2.516	-0.234	-5.310	-1.881	-0.265	-2.198	-0.250
1200	-4.454	-2.699	-0.178	-5.160	-1.992	-0.206	-2.346	-0.192
1300	-4.345	-2.773	-0.130	-5.019	-2.100	-0.151	-2.436	-0.140
1400	-4.239	-2.847	-0.127	-4.778	-2.308	-0.143	-2.578	-0.135
1500	-4.140	-2.916	-0.124	-4.666	-2.390	-0.140	-2.653	-0.132
1600	-4.012	-3.016	-0.080	-4.578	-2.450	-0.092	-2.733	-0.086
1750	-3.946	-3.043	-0.039	-4.534	-2.455	-0.045	-2.749	-0.042
2000	-3.803	-3.128	-0.038	-4.267	-2.664	-0.043	-2.896	-0.040
2500	-3.604	-3.230	-0.036	-4.021	-2.813	0.362	-3.022	-0.038

Δq(e) 0.8

	Sodium			Carbon			Averaged	
Temperature (K)	Log D(cm^2/s)	Log η (Pa.s)	Error	Log D (cm^2/s)	Log_{10} η (Pa.s)	Error	Log_{10} η (Pa.s)	Error
800	-5.491	-1.838	-0.494	-5.947	-1.383	-0.535	-1.610	-0.515
900	-5.238	-2.040	-0.471	-5.821	-1.456	-0.524	-1.748	-0.498
1000	-4.899	-2.333	-0.343	-5.390	-1.842	-0.377	-2.087	-0.360
1100	-4.694	-2.496	-0.235	-5.243	-1.947	-0.262	-2.222	-0.248
1200	-4.491	-2.662	-0.180	-5.050	-2.103	-0.202	-2.382	-0.191
1300	-4.380	-2.739	-0.131	-4.962	-2.156	-0.149	-2.448	-0.140
1400	-4.274	-2.812	-0.128	-4.839	-2.247	-0.145	-2.530	-0.137
1500	-4.203	-2.853	-0.126	-4.709	-2.347	-0.141	-2.600	-0.134
1600	-4.055	-2.973	-0.081	-4.623	-2.405	-0.092	-2.689	-0.087
1750	-3.976	-3.013	-0.040	-4.463	-2.526	-0.045	-2.770	-0.042
2000	-3.822	-3.109	-0.038	-4.287	-2.644	-0.043	-2.876	-0.041
2500	-3.598	-3.236	-0.036	-3.978	-2.856	-0.040	-3.046	-0.038

Δq(e) 1.28

	Sodium			Carbon			Average	
Temperature (K)	Log D(cm^2/s)	Log η (Pa.s)	Error	Log D (cm^2/s)	Log_{10} η (Pa.s)	Error	Log_{10} η (Pa.s)	Error
800	-5.444	-1.885	-0.490	-5.809	-1.520	-0.523	-1.703	-0.506
900	-5.273	-2.005	-0.475	-5.825	-1.453	-0.524	-1.729	-0.499
1000	-4.895	-2.337	-0.343	-5.476	-1.756	-0.383	-2.047	-0.363
1100	-4.659	-2.532	-0.233	-5.146	-2.044	-0.257	-2.288	-0.245
1200	-4.525	-2.628	-0.181	-5.025	-2.128	-0.201	-2.378	-0.191
1300	-4.335	-2.783	-0.130	-4.922	-2.196	-0.148	-2.489	-0.139
1400	-4.221	-2.865	-0.127	-4.743	-2.343	-0.142	-2.604	-0.134
1500	-4.119	-2.937	-0.124	-4.656	-2.400	-0.140	-2.668	-0.132
1600	-4.051	-2.977	-0.081	-4.556	-2.473	-0.091	-2.725	-0.086
1750	-3.940	-3.049	-0.039	-4.471	-2.518	-0.045	-2.783	-0.042
2000	-3.796	-3.136	-0.038	-4.239	-2.692	-0.042	-2.914	-0.040
2500	-3.551	-3.283	-0.036	-4.031	-2.803	-0.040	-3.043	-0.038

Δq(e) 1.76

	Sodium			Carbon			Average	
Temperature (K)	Log D(cm^2/s)	Log η (Pa.s)	Error	Log D (cm^2/s)	Log_{10} η(Pa.s)	Error	Log_{10} η (Pa.s)	Error
800	-5.525	-1.805	-0.497	-5.965	-1.364	-0.537	-1.584	-0.517
900	-5.249	-2.029	-0.472	-5.786	-1.492	-0.521	-1.761	-0.497
1000	-4.886	-2.346	-0.342	-5.516	-1.716	-0.386	-2.031	-0.364
1100	-4.630	-2.560	-0.232	-5.061	-2.129	-0.253	-2.345	-0.242
1200	-4.459	-2.694	-0.178	-4.973	-2.180	-0.199	-2.437	-0.189
1300	-4.327	-2.791	-0.130	-4.833	-2.286	-0.145	-2.539	-0.137
1400	-4.176	-2.910	-0.125	-4.714	-2.372	-0.141	-2.641	-0.133
1500	-4.120	-2.936	-0.124	-4.634	-2.422	-0.139	-2.679	-0.131
1600	-4.004	-3.024	-0.080	-4.492	-2.536	-0.090	-2.780	-0.085
1750	-3.945	-3.044	-0.039	-4.415	-2.574	-0.044	-2.809	-0.042
2000	-3.760	-3.171	-0.038	-4.203	-2.728	-0.042	-2.949	-0.040
2500	-3.550	-3.284	-0.036	-3.966	-2.868	-0.040	-3.076	-0.038

Δq(e) 2.24

	Sodium			Carbon			Average	
Temperature (K)	Log D(cm^2/s)	Log η (Pa.s)	Error	Log D (cm^2/s)	Log_{10} η (Pa.s)	Error	Log_{10} η (Pa.s)	Error
800	-5.396	-1.933	-0.486	-5.810	-1.519	-0.523	-1.726	-0.504
900	-5.058	-2.220	-0.455	-5.568	-1.710	-0.501	-1.965	-0.478
1000	-4.732	-2.500	-0.331	-5.279	-1.953	-0.370	-2.227	-0.350
1100	-4.539	-2.652	-0.227	-5.096	-2.095	-0.255	-2.373	-0.241
1200	-4.356	-2.797	-0.174	-4.811	-2.342	-0.192	-2.570	-0.183
1300	-4.272	-2.846	-0.128	-4.729	-2.389	-0.142	-2.617	-0.135
1400	-4.115	-2.971	-0.123	-4.577	-2.509	-0.137	-2.740	-0.130
1500	-4.058	-2.998	-0.122	-4.519	-2.537	-0.136	-2.768	-0.129
1600	-4.023	-3.005	-0.080	-4.506	-2.522	-0.090	-2.764	-0.085
1750	-3.867	-3.122	-0.039	-4.347	-2.642	-0.043	-2.882	-0.041
2000	-3.748	-3.183	-0.037	-4.197	-2.734	-0.042	-2.959	-0.040
2500	-3.533	-3.301	-0.035	-3.933	-2.902	-0.039	-3.101	-0.037

Δq(e) 2.28

	Sodium			Carbon			Average	
Temperature (K)	Log D(cm^2/s)	Log η (Pa.s)	Error	Log D (cm^2/s)	Log_{10} η (Pa.s)	Error	Log_{10} η (Pa.s)	Error
800	-5.295	-2.034	-0.477	-5.894	-1.435	-0.530	-1.735	-0.503
900	-4.896	-2.382	-0.441	-5.335	-1.943	-0.480	-2.162	-0.460
1000	-4.635	-2.597	-0.324	-5.086	-2.146	-0.356	-2.372	-0.340
1100	-4.461	-2.730	-0.223	-4.958	-2.233	-0.248	-2.481	-0.235
1200	-4.353	-2.800	-0.174	-4.823	-2.330	-0.193	-2.565	-0.184
1300	-4.186	-2.932	-0.126	-4.668	-2.451	-0.140	-2.691	-0.133
1400	-4.089	-2.998	-0.123	-4.543	-2.543	-0.136	-2.770	-0.129
1500	-4.011	-3.045	-0.120	-4.478	-2.578	-0.134	-2.812	-0.127
1600	-3.958	-3.070	-0.079	-4.434	-2.594	-0.089	-2.832	-0.084
1750	-3.835	-3.154	-0.038	-4.347	-2.642	-0.043	-2.898	-0.041
2000	-3.710	-3.221	-0.037	-4.132	-2.800	-0.041	-3.010	-0.039
2500	-3.528	-3.306	-0.035	-3.971	-2.864	-0.040	-3.085	-0.037

Appendix 2.

The mean square displacement used to calculate diffusion and viscosity for the $55K_2CO_3$-$45MgCO_3$ liquids are shown in figure A2 with the individual contributions from K, Mg, C and O shown as a function of temperature.

Selected pair distribution functions obtained directly from the ab initio MD (VASP) simulation trajectories for liquid $55K_2CO_3$-$45MgCO_3$ are shown in figure A3 for different temperatures and shown the evolution of the liquid structure. These partial contributions are shown for four temperatures and demonstrate the change in the local environment of potassium as a function of temperature with a broader distribution at higher-temperature and an increase in coordination number and formation of better defined edge-shared K-O coordination polyhedra as temperature is reduced (Figs. A3a and A3b). The C-O coordination environment remains unchanged with temperature (Fig. A3c) as expected and although there is a broadening of the C-C contribution (Fig. A3d), there is no evidence for the development of a second C-C distance that characterizes chain formation in Na_2CO_3 (Wilson et al. 2018). The changes in O-O (Fig. A3e) and the shift to lower radial distance with temperature for the Mg-Mg contribution (Fig. A3f) all indicate that the structural changes with temperature that are reflected in the fragile nature of the $55K_2CO_3$-$45MgCO_3$ liquid are associated with the changes in the local environment of potassium.