

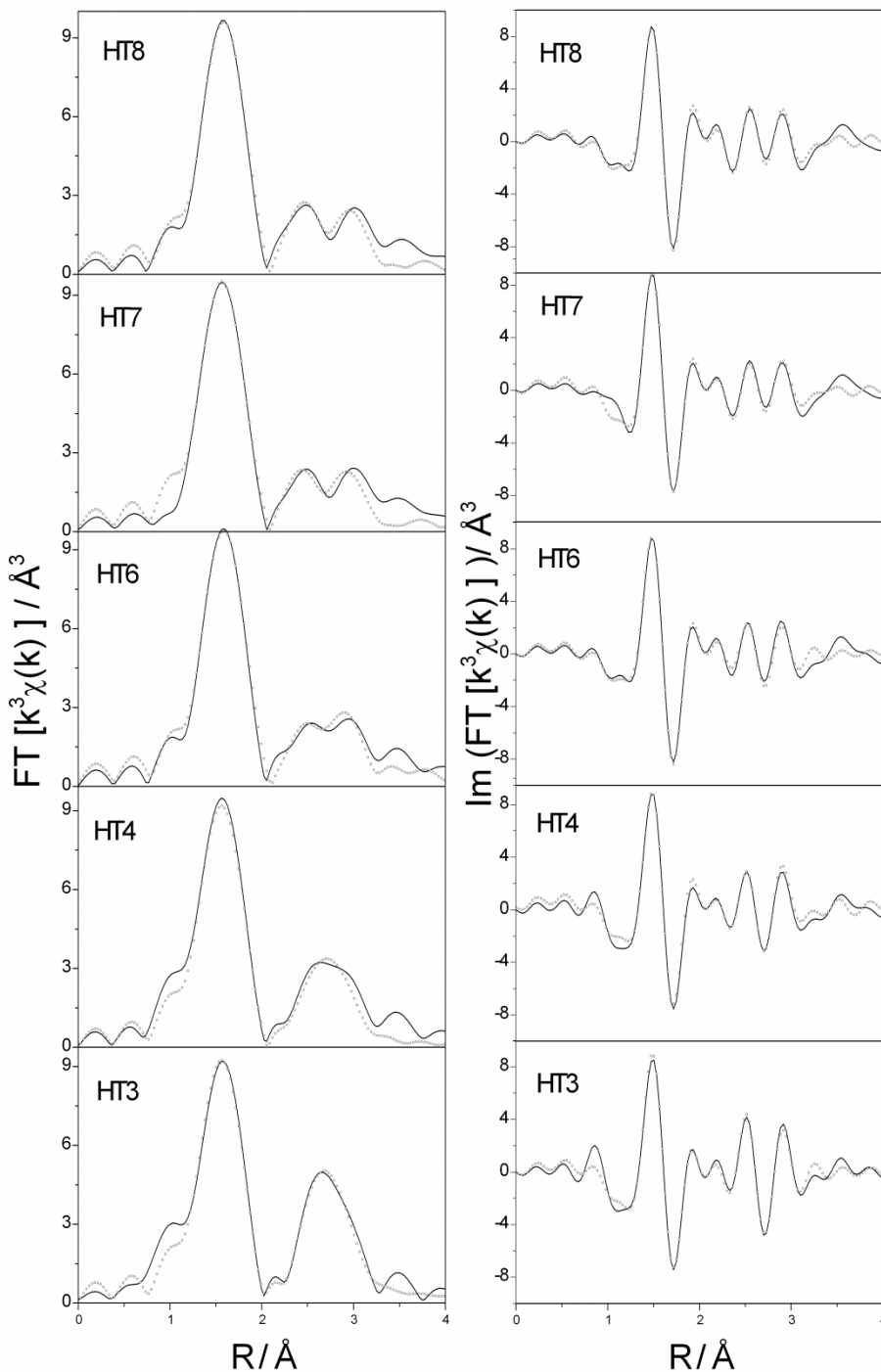


Figure S1 - Fourier-transformed magnitude and imaginary parts of the experimental and simulated spectra for NiAl-TA hydrotalcites. Left, Fourier Transform Magnitude; Right, Fourier Transform Imaginary Part; solid line, experimental spectra; dotted line, simulation

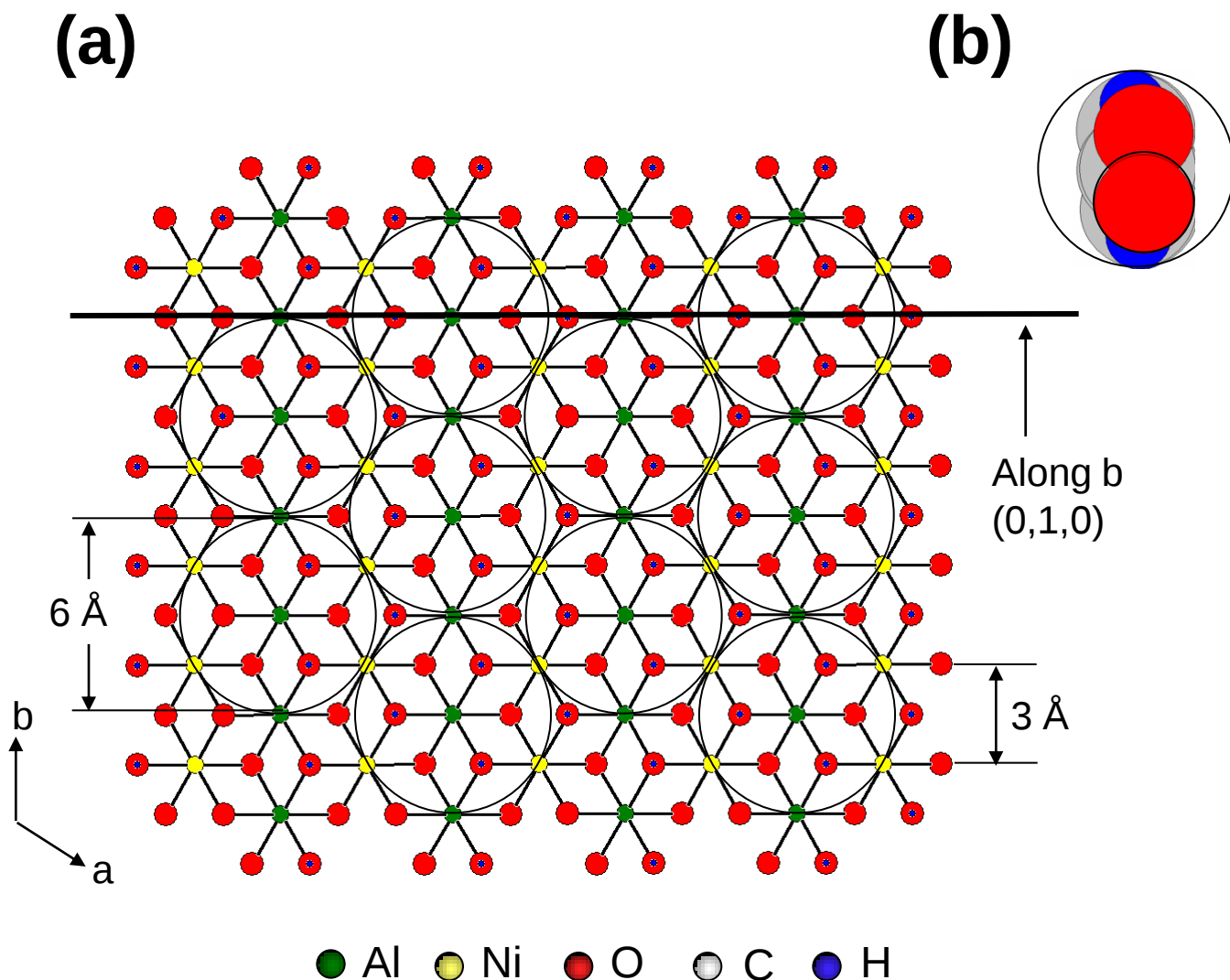


Fig. S2 – (a) Model view of a Ni-Al-terephthalate layer with $x = 0.5$ along the c axis. The circles have the same diameter as the one in Fig. S4(b). Figure drawn using crystallographic data for Mg-Al- CO_3^{2-} hydrotalcite from Inorganic Crystal Structure Database (ICSD) code 81963; (b) Space-filling view of the terephthalate anion along the carboxyl axis. Figure drawn using crystallographic data for cobalt terephthalate from James A. Kaduk, *Acta Cryst.* 2002, **B58**, 815-822. The circle represents the area swept by a freely rotating anion. Van der Waals radii taken as 1 Å for H, 1.77 Å for C and 1.52 Å for O. The cut along the b axis is shown in Fig. S3.

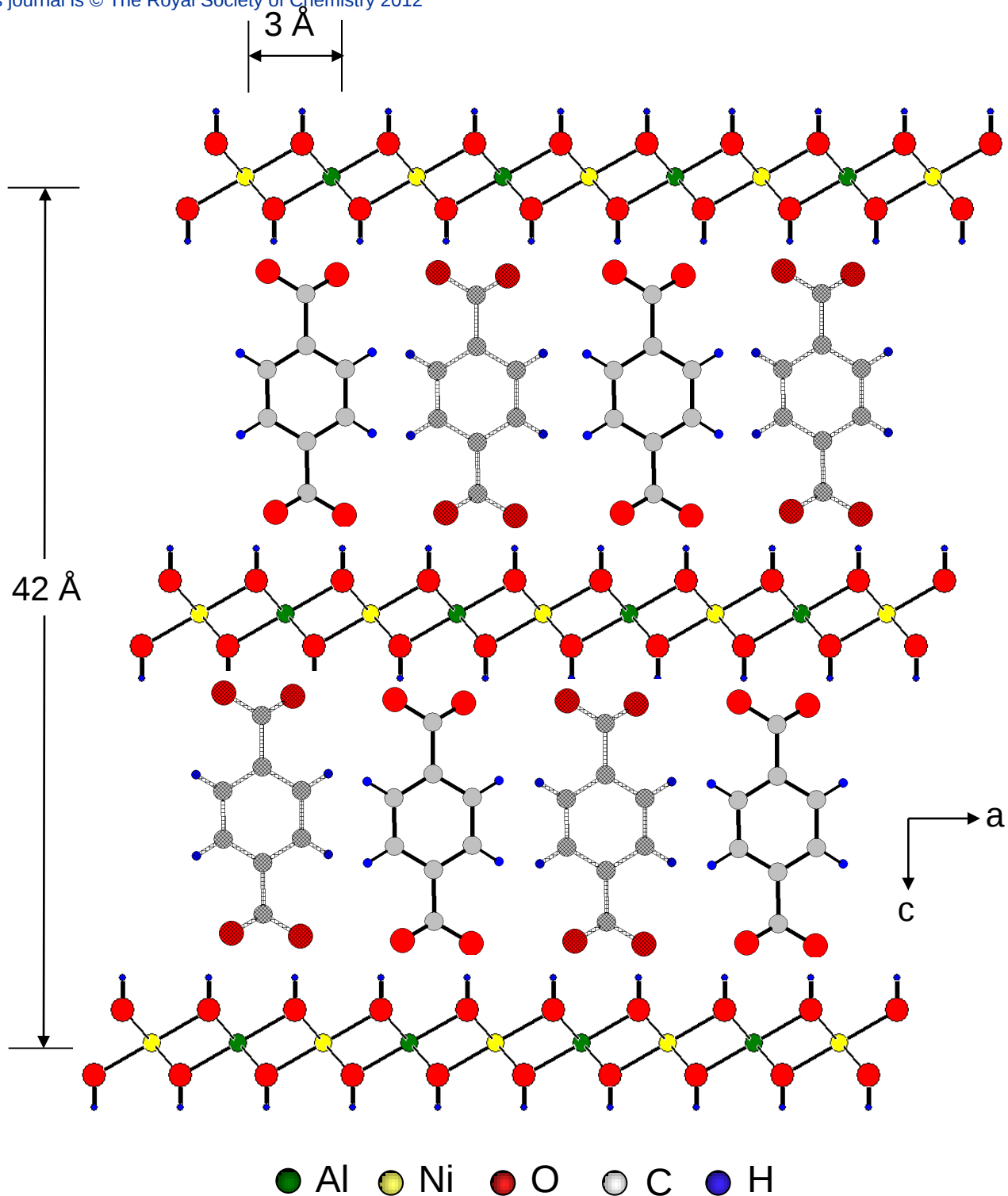


Fig. S3 – Layer chart for Ni-Al-terephthalate HDL with $x = 0.5$ along the b axis. Figure based on the same crystallographic data for each layer and for the terephthalate anion as in Fig. S2. Shaded terephthalate anions are at the background plane and unshaded ones at the foreground plane.