Supporting Information (SI)

"Quantitative correlation between the protein primary sequences and secondary structures in spider silks" by J.E. Jenkins *et al.*

$^{1}H \rightarrow ^{13}C$ CP and ^{13}C DD-MAS spectra with a rapid recycle delay were collected of Ma and Mi silk in their dry and wetted state on a Varian VNMRS 400 MHz wide-bore spectrometer equipped with a 3.2 mm triple resonance probe operating in triple resonance mode ($^{1}H/^{13}C/^{15}N$). $^{1}H \rightarrow ^{13}C$ CP were collected at 5 kHz MAS matched on the Hartmann-Hahn condition with a 3.4 μs ^{1}H pulse, a CP rf field strength of 75 kHz with a 1 ms contact time, and ~100 kHz TPPM ^{1}H decoupling. ^{13}C DD-MAS were collected at 20 kHz MAS with a 3 μs ^{13}C pulse, a recycle delay of 1 s, and ~100 kHz TPPM ^{1}H decoupling.

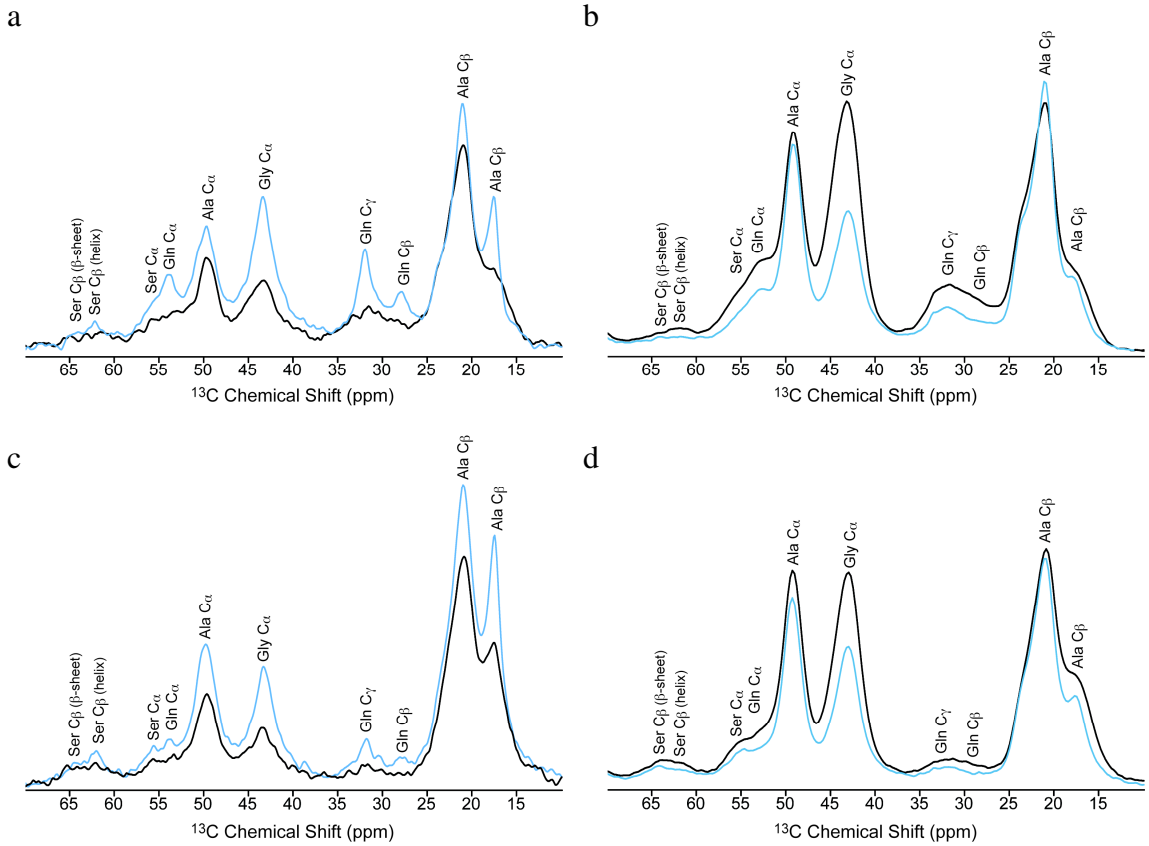

Figure S1 | ^{13}C DD-MAS of N. clavipes (a) Ma and (c) Mi silk in the water wetted state (blue) and dry (black). ^{13}C CP-MAS of (b) Ma and (d) Mi silk in the dry state (black) and wet state (blue). The Ser C_β in a helical conformation is enhanced in the ^{13}C DD-MAS when silk is wetted due to an increase in mobility however, this increase in mobility cause CP to be inefficient and the helical Ser C_β to decrease in intensity in the ^{13}C CP-MAS. ^{13}C DD-MAS and ^{13}C CP-MAS were collected of Ma and Mi in the dry and wetted state to determine that the upfield (lower ppm) side of the broad Ser C_β peak is in a mobile helical conformation and the downfield (higher ppm) shoulder is from the Ser C_β that adopts a rigid β-sheet secondary structure.