

**Figure 1 Supplemental:** REDOR dephasing curve for statherin and mutants adsorbed onto HAP, hydrated. Error bars are omitted for clarity, but were typically 5-15%. stath(WT) (●), stath(K6A) (▲), stath(R9A) (■), stath(R10A) (▲), stath(R13A) (●) and stath(KRA) (◆). The data are best simulated by fits of 4.2 Å (red line), 4.3 Å (pink line), 4.4 Å (brown), 4.5 Å (green) and 4.6 Å (blue), with the fit associated with the data points of the associated color.

**Figure 2 Supplemental:**  $T_{1\rho}$  data of the mutants and wild type protein, frozen and at RT. Each mutant is shown under both conditions. The average of the  $T_{1\rho}$  (24.1 ms) of all of the frozen proteins (■) and the average of the  $T_{1\rho}$  (9.9 ms) of all of the RT proteins (●) is also shown with error bars (2-5%), indicating the increase in mobility of the protein under mobile conditions (RT), but emphasizing the very small change from mutant to mutant under frozen and RT conditions. Inset:  $^{13}\text{C}$  NMR spectra of stath(WT) frozen (top) is shown for a comparison of an immobile CSA and hydrated (bottom) as a typical example of the hydrated, RT samples.