

Supplementary Material

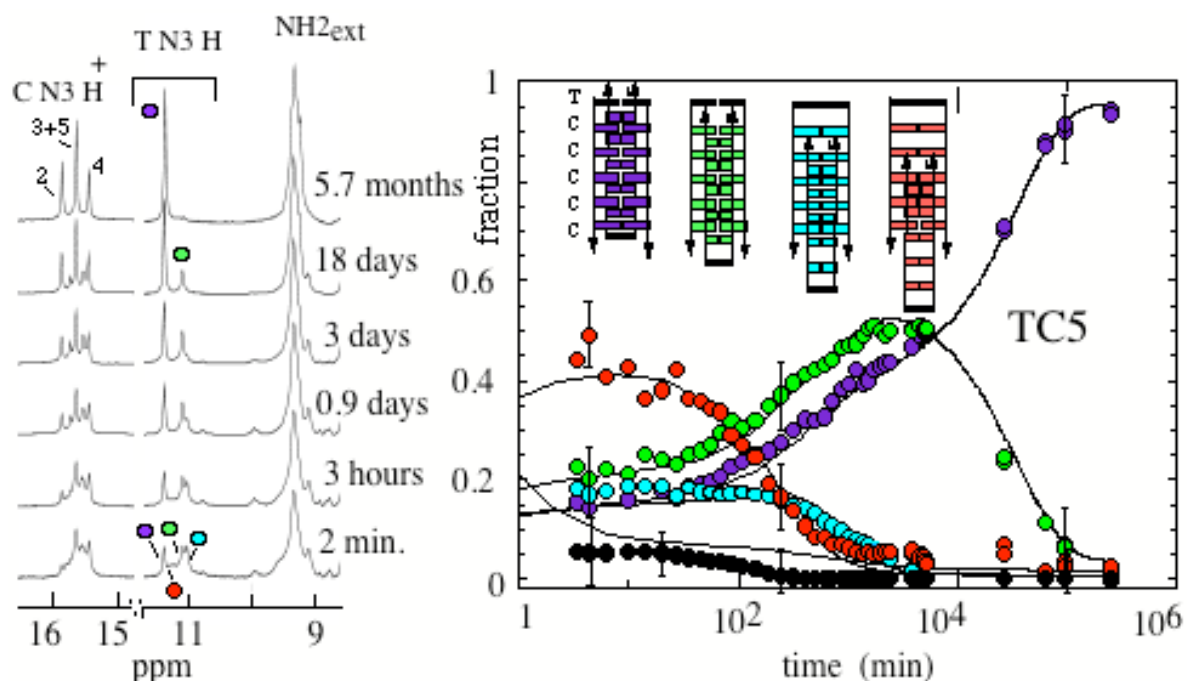


Fig. S1

Figure S1. Association of TC5 into fully and partially intercalated i-motif tetramers at 0 °C. Left panel: Selected spectra of a 0.1mM TC5 solution initially monomeric collected after incubation during the times indicated on the figure. The spectra show the formation of at least four tetramers whose T imino protons are indicated by colored circles. The fully intercalated tetramer (purple) is identified by the spectrum at equilibrium (1\$). Assuming that the tetramers lifetimes are proportional to the number of intercalated C•C+ pairs, we tentatively identified each tetramer with the species displayed on the right panel. Right panel: Evolution of the monomer (black circle) and tetramer fractions. The solid lines are computed according to a system of four parallel monomer-tetramer reactions. The lifetime of the fully intercalated species is estimated longer than $3 \cdot 10^5$ min. in agreement with fig 1@. The lifetimes of the green (7500 min.), cyan (250min.) and red species (60 min.) were adjusted to get the best fits. The

half formation time of the fully and partially intercalated species are not significantly different: $\tau=0.28\pm0.1$ min.