

**Supplementary Table 1.** Residue-specific chemical shift differences ( $\Delta\omega$ ) between the ground and excited states of yAAC3 determined from the fitting of relaxation dispersion data to a two-state exchange model

Residue Number	Free yAAC3 $^{15}\text{N}$ $\Delta\omega$ (ppm)	yAAC3-ADP $^{15}\text{N}$ $\Delta\omega$ (ppm)	yAAC3-CATR $^{15}\text{N}$ $\Delta\omega$ (ppm)
21	1.0 $\pm$ 0.5	no data	no data
26	5.7 $\pm$ 0.9	5.9 $\pm$ 0.4	6.1 $\pm$ 1.0
27	3.5 $\pm$ 0.7	2.2 $\pm$ 1.4	2.6 $\pm$ 0.8
30	3.6 $\pm$ 1.1	2.9 $\pm$ 1.5	no data
31	5.5 $\pm$ 1.4	5.3 $\pm$ 1.1	5.7 $\pm$ 1.4
41	3.1 $\pm$ 0.2	3.7 $\pm$ 1.0	4.6 $\pm$ 0.8
42	4.6 $\pm$ 0.6	4.0 $\pm$ 1.0	no data
61	4.7 $\pm$ 0.5	3.2 $\pm$ 1.2	3.1 $\pm$ 0.3
70	2.2 $\pm$ 0.3	1.8 $\pm$ 1.3	1.2 $\pm$ 0.4
78	1.3 $\pm$ 0.2	no data	no data
81	1.2 $\pm$ 0.2	1.4 $\pm$ 0.4	no data
82	2.5 $\pm$ 0.7	1.8 $\pm$ 0.9	1.5 $\pm$ 0.2
84	3.7 $\pm$ 1.2	4.2 $\pm$ 1.3	5.1 $\pm$ 1.2
85	4.9 $\pm$ 1.2	7.8 $\pm$ 1.5	no data
91	1.2 $\pm$ 0.2	1.3 $\pm$ 0.5	4.9 $\pm$ 0.5
95	2.1 $\pm$ 0.8	2.0 $\pm$ 1.1	2.7 $\pm$ 0.6
96	3.2 $\pm$ 0.3	no data	no data
124	1.2 $\pm$ 0.2	no data	no data
146	1.1 $\pm$ 0.6	0.3 $\pm$ 0.2	no data
166	1.1 $\pm$ 1.0	0.8 $\pm$ 0.6	no data
167	no data	1.1 $\pm$ 0.2	no data
170	1.5 $\pm$ 0.6	no data	no data
174	1.3 $\pm$ 0.3	1.1 $\pm$ 0.3	no data
246	2.4 $\pm$ 0.2	1.6 $\pm$ 0.2	1.5 $\pm$ 0.3
247	1.2 $\pm$ 0.6	0.9 $\pm$ 0.5	0.7 $\pm$ 0.5
268	1.4 $\pm$ 0.3	1.1 $\pm$ 0.3	0.6 $\pm$ 0.5
291	5.5 $\pm$ 1.4	no data	5.3 $\pm$ 0.4
292	not resolved	not resolved	6.2 $\pm$ 0.4
294	4.5 $\pm$ 0.2	4.2 $\pm$ 1.0	no data

**Supplementary Table 2.** Kinetic and thermodynamic parameters of yAAC3 derived from relaxation dispersion measurements measured at three fields.

State	Protein Region	$k_{ex}$ (s <sup>-1</sup> )	$p_e$ (%)
Free yAAC3	TM helices	470 ± 100	1.5 ± 0.2
yAAC3-ADP	TM helices	1600 ± 300	2.2 ± 0.5