

Complete ion-coordination structure in the rotor ring of Na⁺-dependent F-ATP synthases

SUPPLEMENTAL DATA

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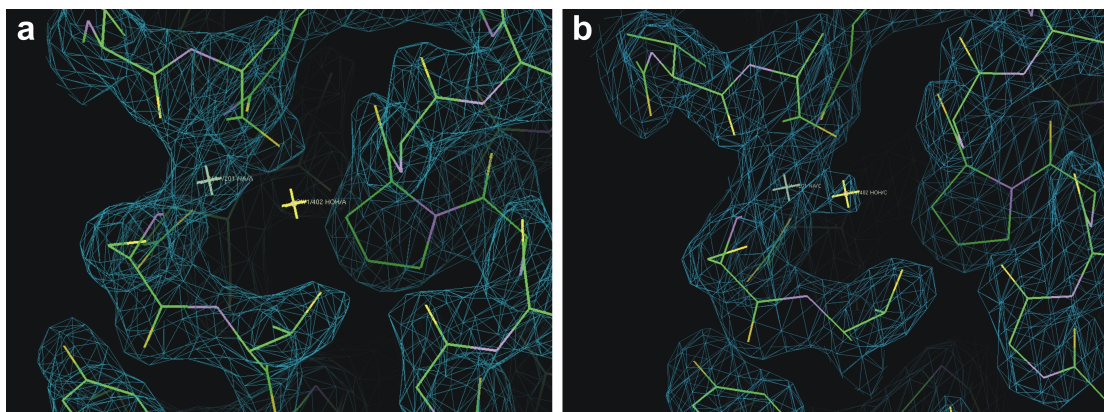


Fig. S1. Electron densities in the *Ilyobacter tartaricus* c-ring.

The electron density peaks ($2F_{\text{obs}} - F_{\text{calc}}$ map at 1.8σ) as observed in¹ are shown. The positions of Na^+ and the water are marked with a grey and orange cross, respectively. Exemplified are in (a) chain A and in (b) chain C.

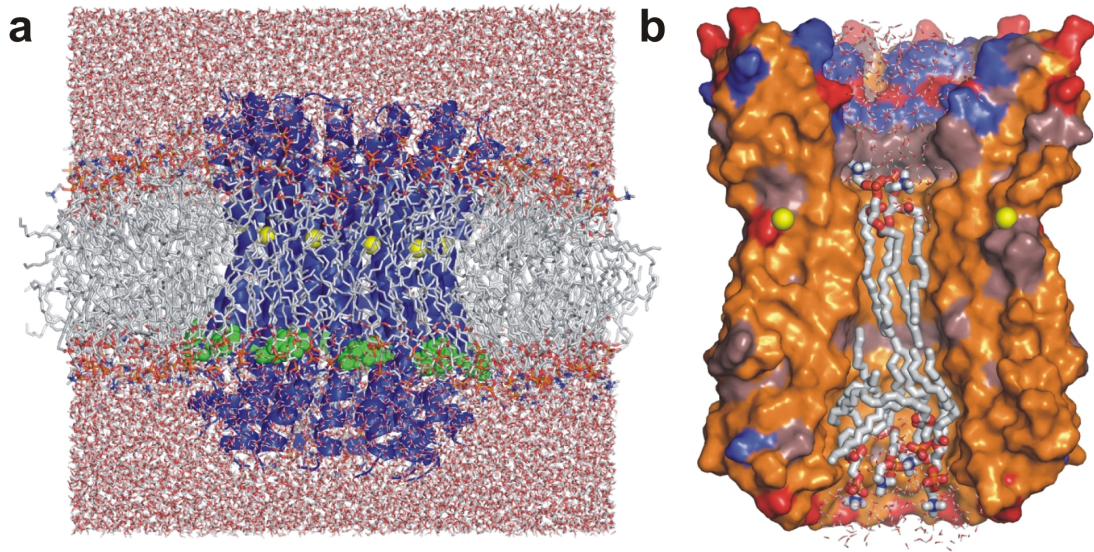


Figure S2 All-atom simulation model of the c-ring of the F₁F₀-ATP synthase from *Ilyobacter tartaricus*, embedded in a POPC lipid membrane.

(a) View along the membrane plane, with the cytoplasmic side at the top. The system comprises 100,754 atoms, *i.e.* 11 c-subunits (*blue*), with 11 bound Na⁺ ions (*yellow*), 237 POPC lipids and 18,353 water molecules (~100,000 atoms). Residue Tyr⁸⁰ (*green*) delineates the water-membrane interface. The periodic simulation box is approximately 100 × 100 × 97 Å³. (b) Close-up of the interior of the ring. Note the asymmetric distribution and number of lipids (*sticks*), which closely match the hydrophobic surface of the protein interior (*orange*), allowing for a wide, water-filled cavity lined by polar residues (*red, blue*) on the cytoplasmic side. This is consistent with AFM images of a homologous ring from *S. elongatus*². During the course of a 15 ns simulation, the mean displacement of the protein backbone, relative to the crystal structure, was 0.50 ± 0.05 Å for individual subunits, and 0.74 ± 0.04 Å as a whole. The mean Na⁺ coordination distances during the simulation are 2.17 ± 0.07 Å for Glu⁶⁵(Oε2) and 2.3 ± 0.1 Å for the other 4 ligands.

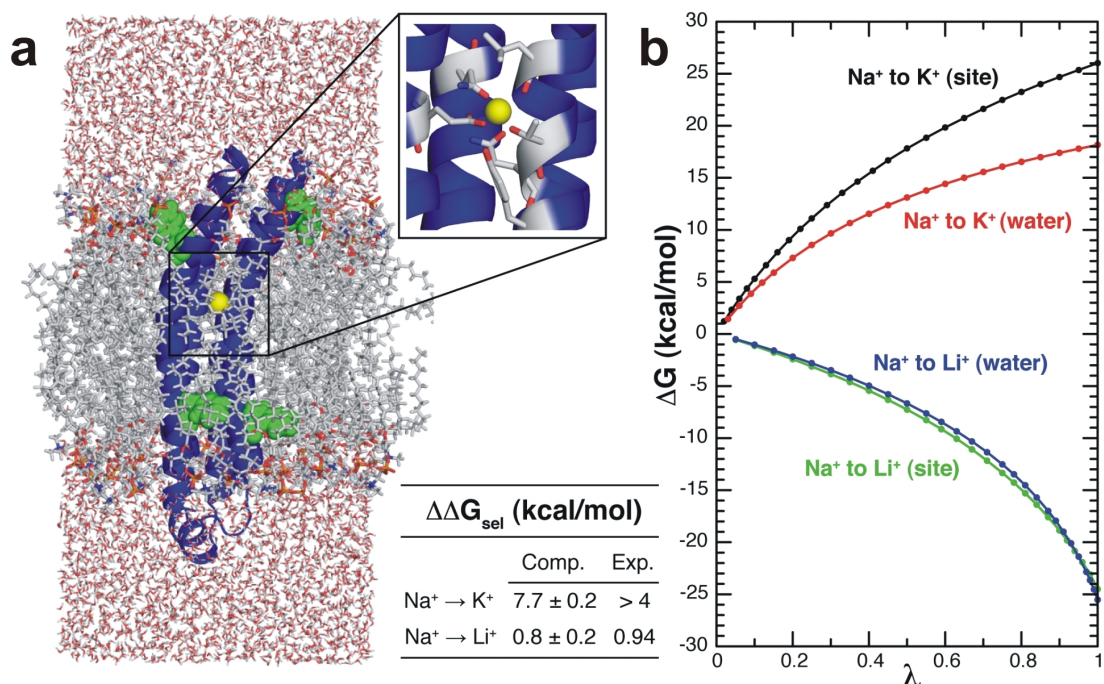


Figure S3. Calculations of the selectivity of ion binding in a K-ring monomer of the V-type ATPase from *Enterococcus hirae*.

(a) All-atom simulation model of a K-ring monomer³, embedded in a POPC membrane. This model was used to benchmark the parameter set subsequently used for the *I. tartaricus* c-ring. The system comprises 23,921 atoms, *i.e.* a 4-helix bundle (*blue*), with a bound Na^+ ion (*yellow*), 60 POPC lipids and 4,523 water molecules. Residue Tyr⁴ and Phe¹³⁴ (*green*) delineate the water-membrane interface. The periodic simulation box is approximately $50 \times 50 \times 97 \text{ \AA}^3$. (b) Free-energy change during the transformation of Na^+ into K^+ or Li^+ , as function of the scaling parameter λ , for either bulk water or the binding site. For clarity, only the forward transformation is shown. Protein and water parameters correspond to the CHARMM27 forcefield and the TIP3P model, respectively⁴. The Lennard-Jones parameters for K^+ and Na^+ are from⁵, that is, for K^+ , $\epsilon = -0.0870$, $\sigma/2 = 1.7637$; and for Na^+ , $\epsilon = -0.0469$, $\sigma/2 = 1.4197$ (ϵ in kcal/mol and σ in \AA). For Li^+ , we used $\epsilon = -0.00932$, $\sigma/2 = 1.1175$ to reproduce the experimental hydration free energy (-25.2 kcal/mol relative to Na^+) under our simulation conditions. In addition, a correction of the Li^+ interaction with the carboxylate and carbonyl oxygen atoms was required in order to reproduce the experimental selectivity of the K-ring⁶. This correction is introduced in the form of an increased σ value for these specific atom pairs, namely $\Delta\sigma = +0.18 \text{ \AA}$. The resulting free-energies of selectivity are compared with experimental values⁶, derived using $\Delta G_{\text{sel}} = -k_{\text{B}}T \log K_{\text{d}} / K_{\text{i}}$.

References

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