

Figure S1. Structural characterization of the S-Layer of *S. acidocaldarius*: Transmission electron microscopic picture (A), Atomic Force microscopic picture (B), and a schematic picture of the cell envelope (C). The outermost SlaA layer possesses p3-symmetry and is fastened to the archaeal cells via another, integrated into the cytoplasmic membrane, anchoring protein, called SlaB.

Reference:

Sonja-Verena Albers, Zalán Szabó & Arnold J. M. Driessen (2006) Protein secretion in the Archaea: multiple paths towards a unique cell surface. *Nature Reviews Microbiology* 4, 537-547

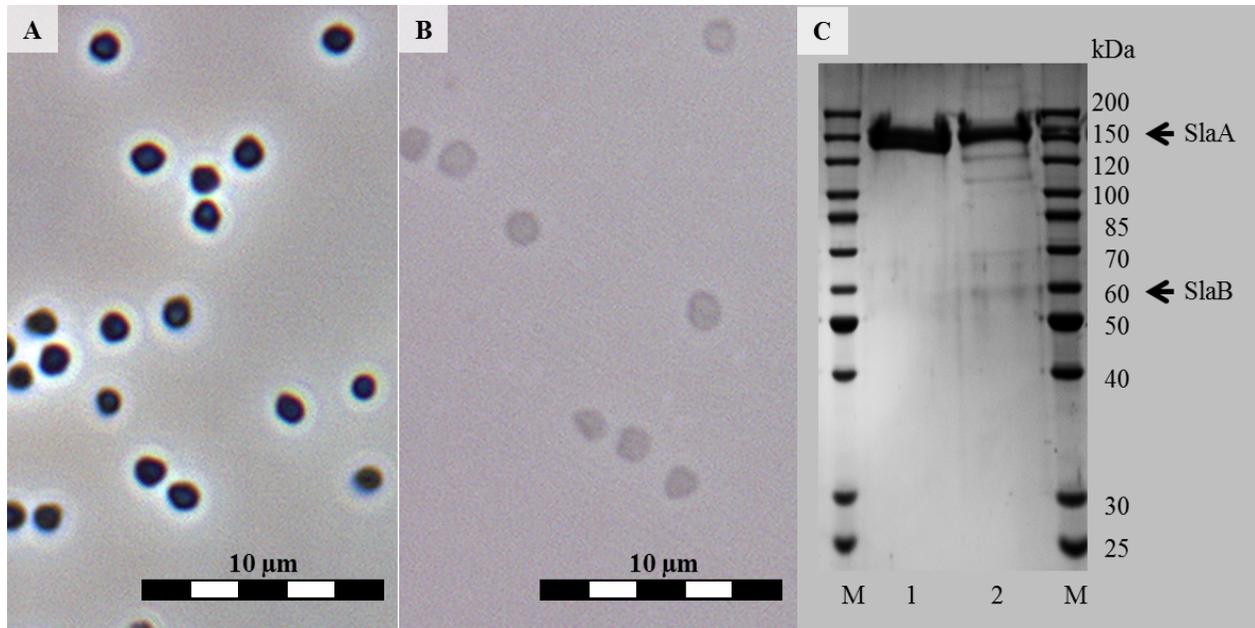


Figure S2. Morphological and biochemical characterization of the S-Layer ghosts: A) Light microscopic pictures of the whole cells of *S. acidocaldarius* and B) of the S-Layer-ghosts - pictures were taken at 1000-fold magnification in phase contrast mode - C) SDS-PAGE protein gel stained with colloidal Coomassie Brilliant Blue G-250: 1) 15 μg of the S-layer ghosts after final purification step with hot SDS only containing the SlaA protein; 2) 10 μg of the partially purified S-layer ghosts, before the final purification step, containing SlaB and other proteins of the cytoplasmic membrane; M) protein marker - PAGERULER™ SM0661 (Fermentas GmbH, Sankt Leon-Rot, Germany).

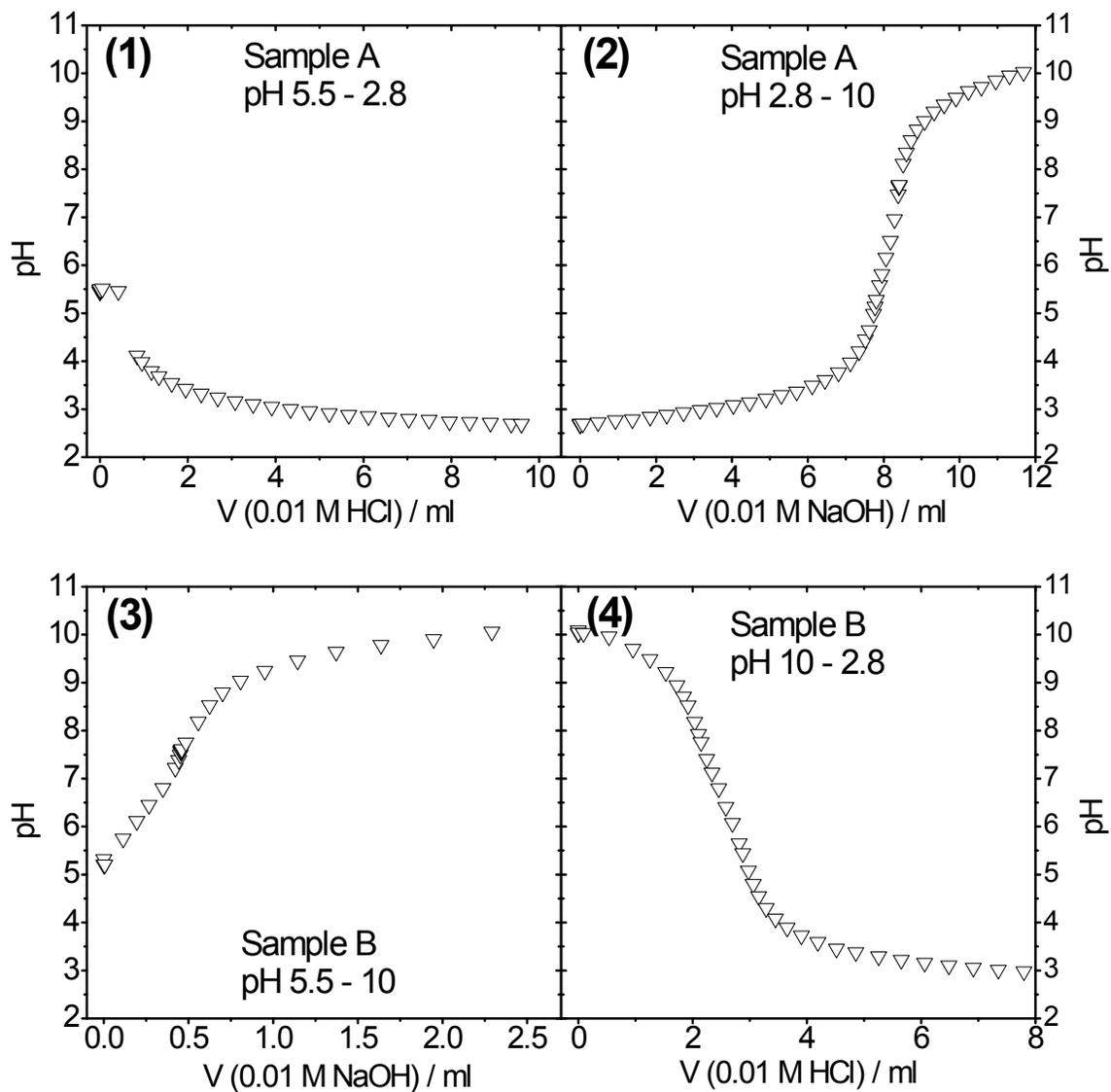


Figure S3. Potentiometric titration of S-layer proteins. (1) Sample A from pH 5.5 down to pH 2.8; (2) same sample (A after titration step 1) from pH 2.8 up to pH 10; (3) sample B from pH 5.5 up to pH 10; (4) same sample (sample B after titration step 3) from pH 10 down to pH 2.8.

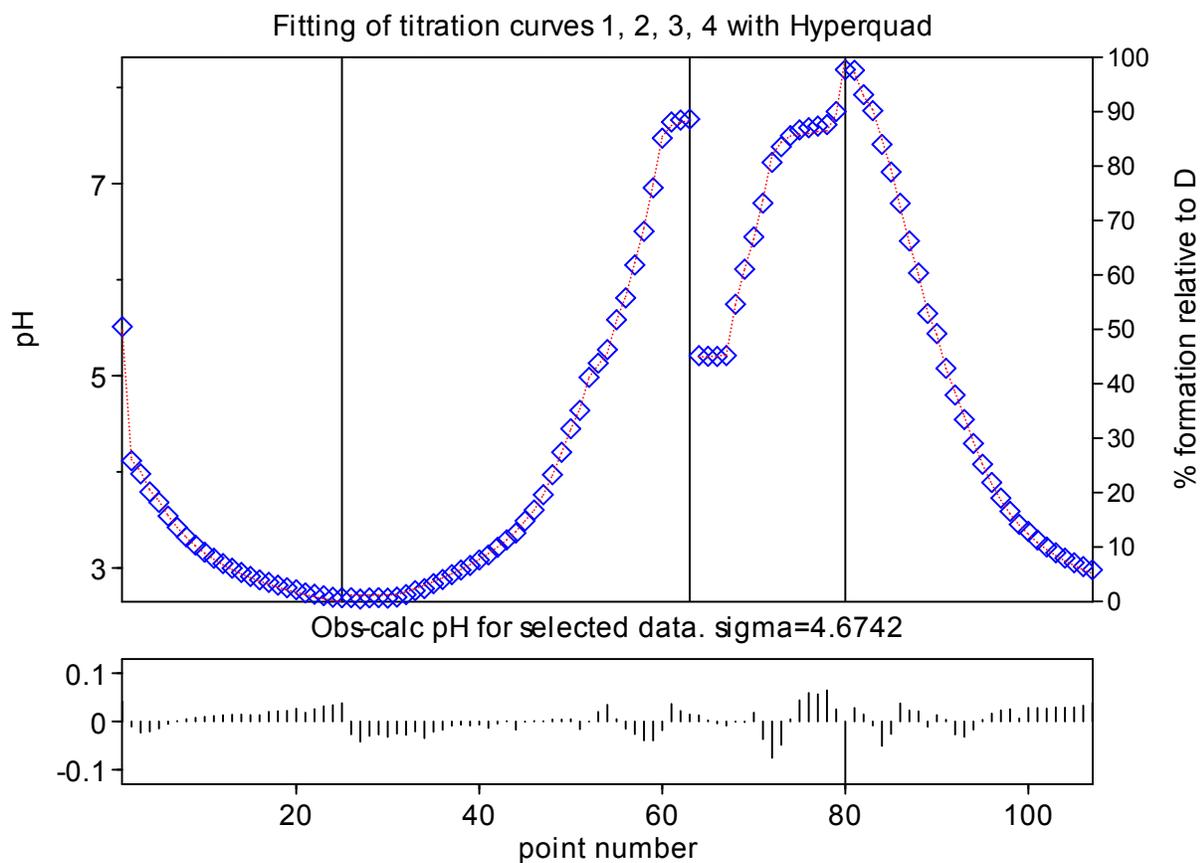


Figure S4. Fitting of the titration data 1,2 (sample A) and 3,4 (sample B). Square: measured data points; dotted line: best fit, calculated by using the software “Hyperquad”. Bottom: corresponding residual.

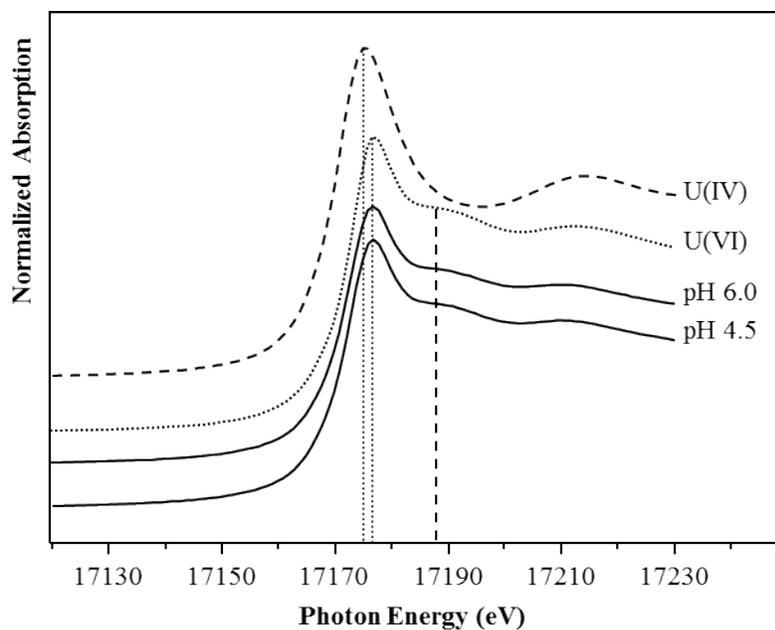


Figure S5. Uranium L_{III} -edge X-ray absorption spectra recorded from the uranium complexes formed at the S-layer of *S. acidocaldarius* at pH 4.5 and 6 together with those of two reference solutions, one of U(VI) and another one of U(IV). The stock solution of U(VI) was obtained by dissolving $\text{Na}_2\text{U}_2\text{O}_7 \times 6 \text{H}_2\text{O}$ in 7 M HClO_4 . Part of this solution was reduced electrochemically to U(IV) at a mercury pool cathode. The uranium oxidation state in these solutions was confirmed by UV/Vis spectroscopy. For comparison, the positions of the white line of U(IV) and U(VI) are illustrated by dotted lines. The position of the absorption peak (~ 17188 eV), which represents the multiple scattering path of the axial oxygen atoms of U(VI) is marked by a dashed line.