Supplementary Material (ESI) for Dalton Transactions

Complexation of uranium(VI) with peptidoglycan

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Electronic supplementary information (ESI)

List of Figures

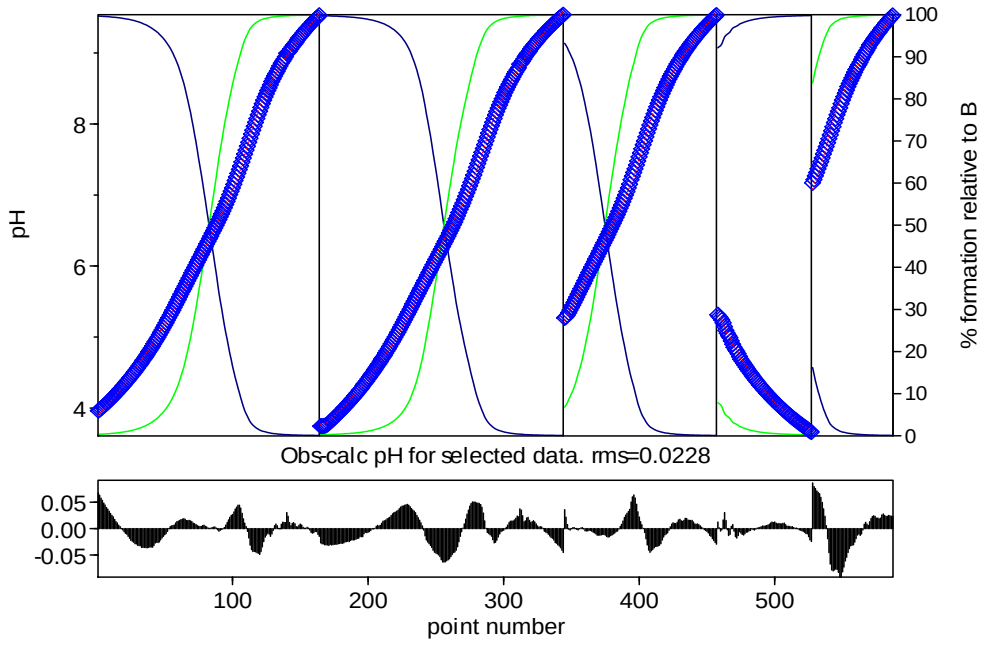

Figure S1: Potentiometric titration curves of PG (blue squares) with fit (red lines) and residuals (lower diagram) from analysis with Hyperquad2006.

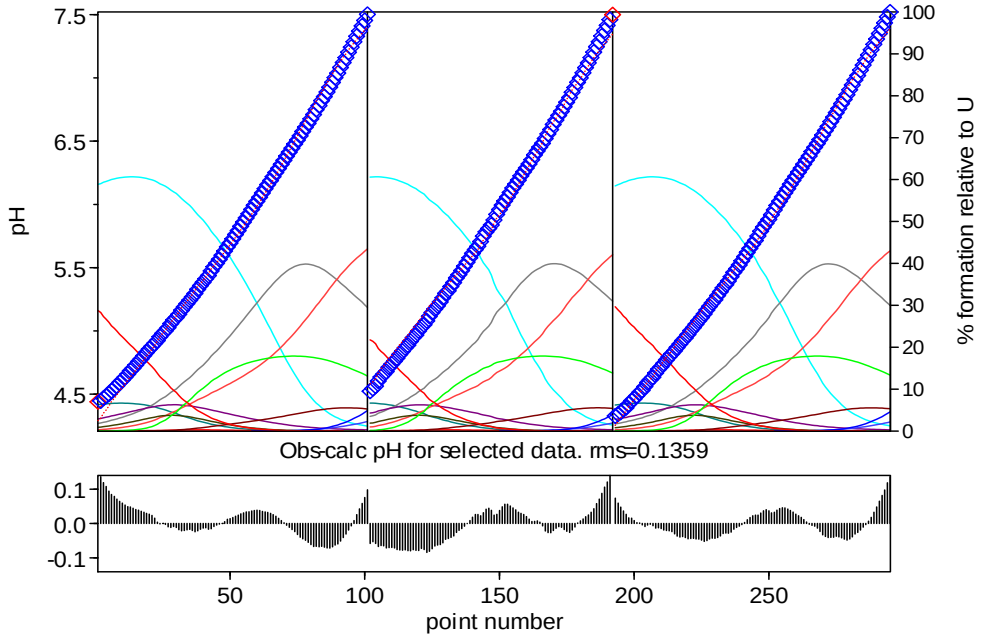

Figure S2: Potentiometric titration curves of PG with uranyl (blue squares) with fit (red lines) and residuals (lower diagram) from analysis with Hyperquad2006.

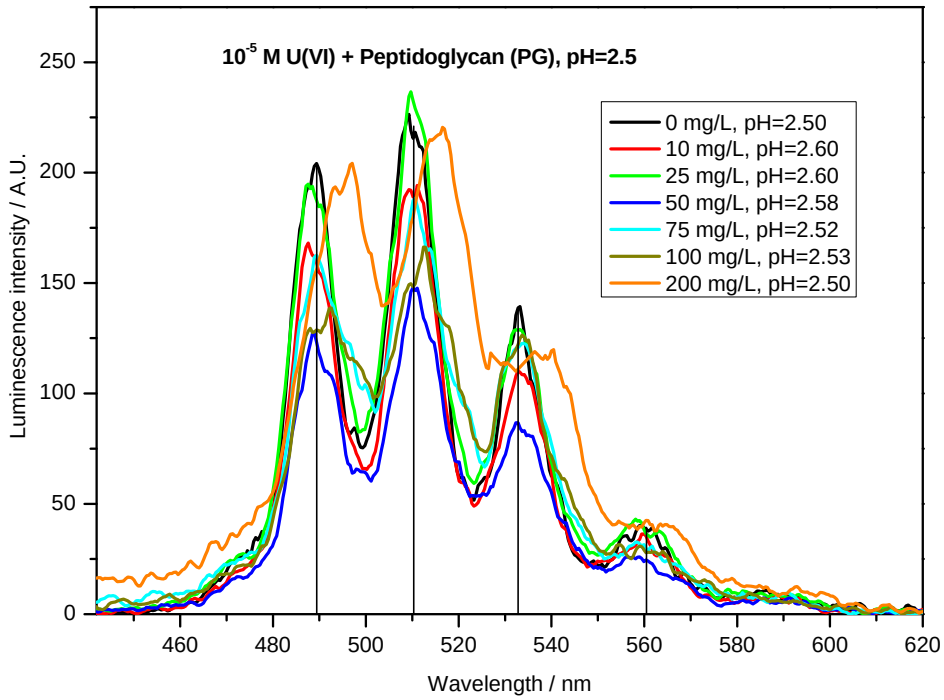

Figure S3: Luminescence spectra of 10^{-5} M U(VI) at pH = 2.5 in dependency of the PG concentration.

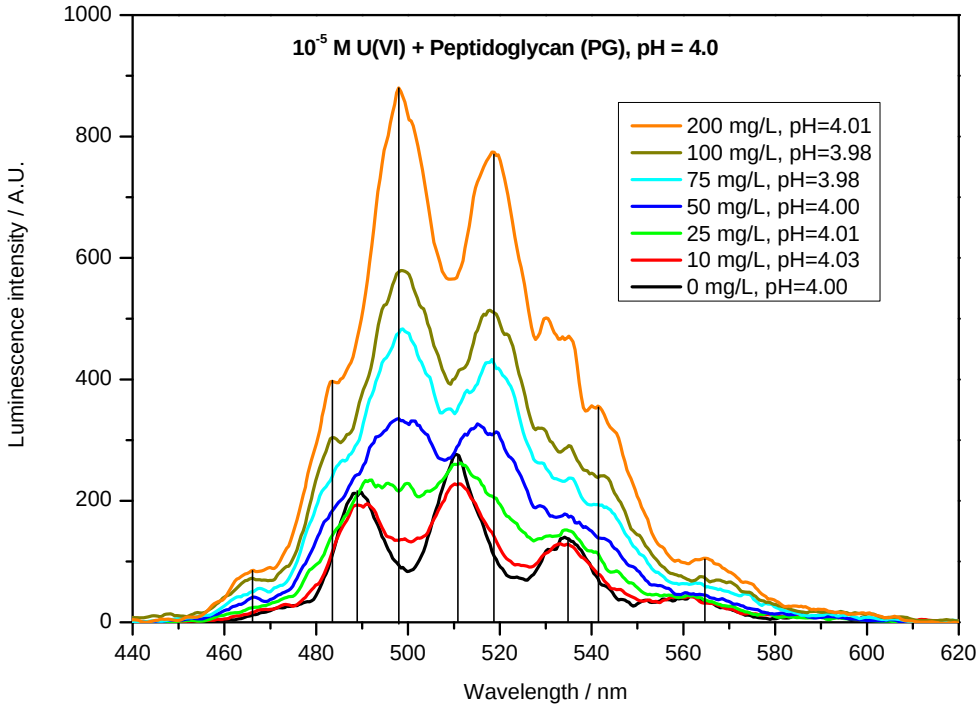

Figure S4: Luminescence spectra of 10^{-5} M U(VI) at pH = 4.0 in dependency of the PG concentration.

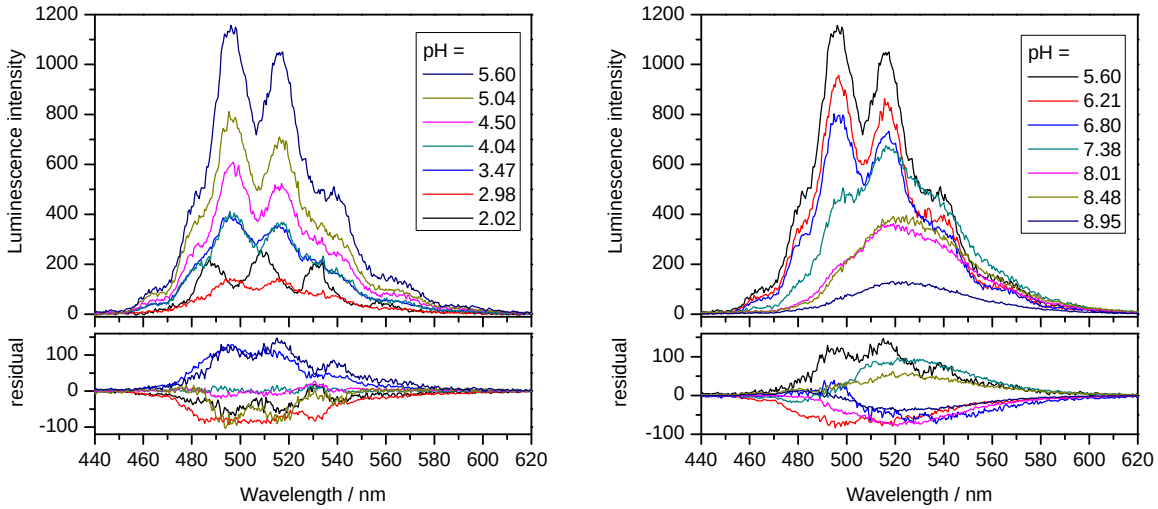

Figure S5: Luminescence spectra of 10^{-5} M U(VI) and 0.1 g/L PG in dependency of pH with the residuals of the analysis with SPECFIT.

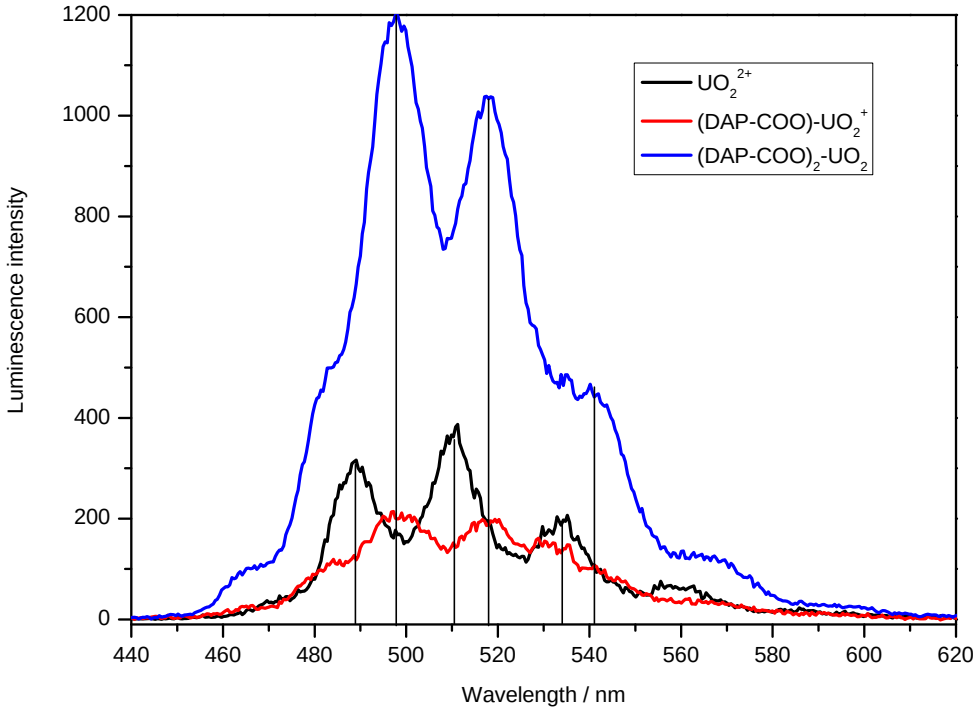

Figure S6: Single spectra of the U(VI) PG complexes, determined with SPECFIT. DAP = Diaminopimelic acid site of PG.

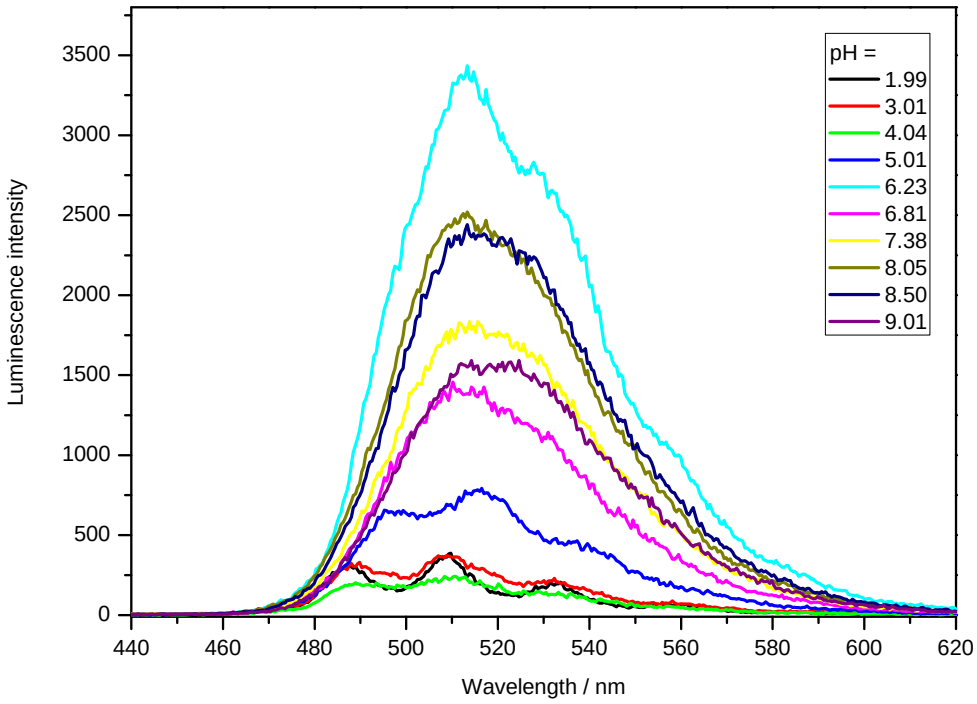

Figure S7: Luminescence spectra of 10^{-5} M U(VI) without PG and CO_2 free in dependency of pH.

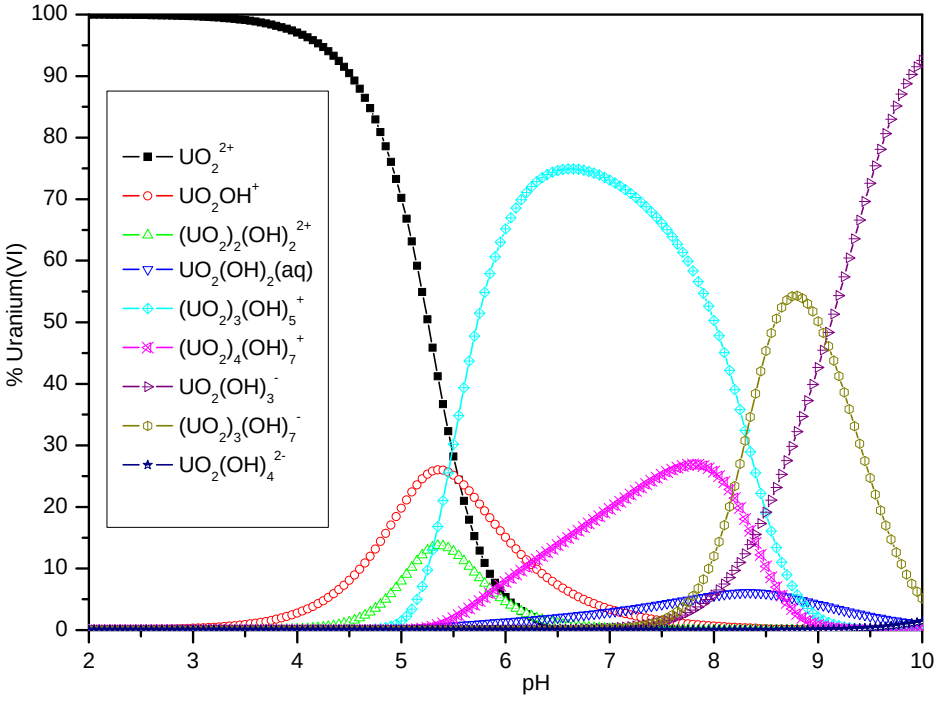

Figure S8: Speciation of 10^{-5} M U(VI) under inert gas conditions (calculated with EQ3/6).

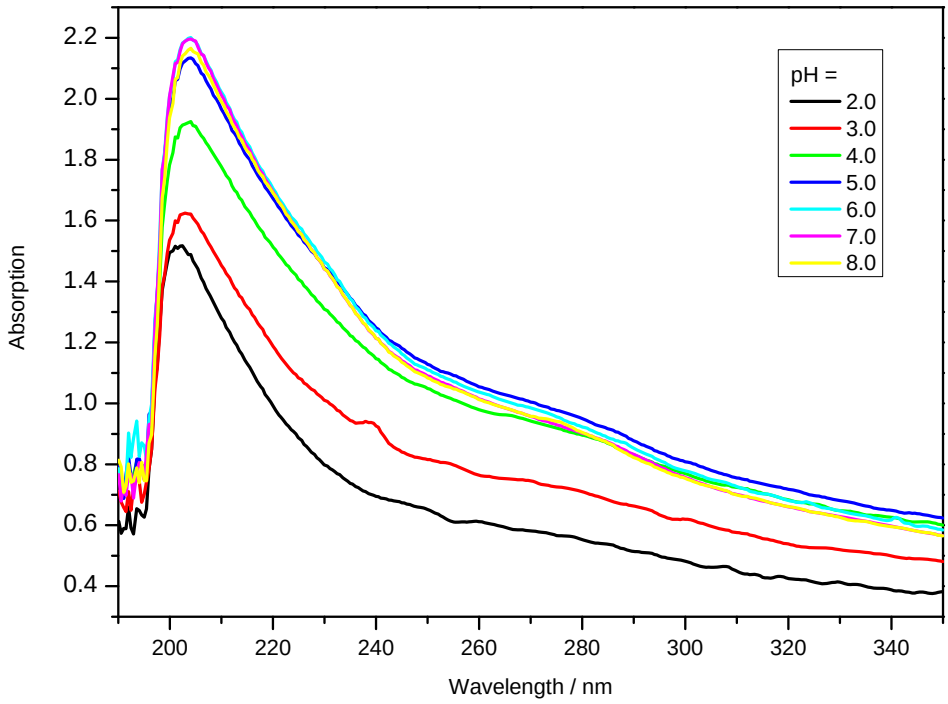

Figure S9: Selected absorption spectra of 0.1 g/L PG in dependency of pH.

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Electronic supplementary information (ESI-2)

Comment on potentiometric data

The discrepancies in the pK_a values of peptidoglycan in the old and new manuscript are not due to the measurement data and not due to the old and new program codes of Hyperquad. It is originated in different ways of determination.
The new program version (Hyperquad 2006) has a big advantage. It provides the possibility to fit all titration curves simultaneously. This increases the precision a lot and makes it possible to fit species with lower formation ratio. The old version (Hyperquad 2000) allowed only the separate fitting of the single titration curves which made the whole evaluation much more imprecisely.
The old data (Hyperquad 2000) were not determined with all measurement data. They were determined with only the two titration curves over the whole pH range (3 to 10), the titration curve from pH 5 to 3 for the first pK_a, and the titration curves from pH 5 to 10 only for the third pK_a. The single determination of the titration curves in the range of pH 5 to 10 resulted in only two pK_a, one pK_a value between 4.9 and 5.8 (probably "averaged" values between the first and second pK_a) and another between 9.5 to 10.0. It was not possible to separate the first pK_a into two values, probably because of the higher starting pH. So we decided to omit the first pK_a´s for calculating the average values and used only the pK_a in the alkaline pH range.
This should be the explanation why the averaged values from the simultaneous fitting of all 5 titration curves with the new version Hyperquad 2006 differ from the old values. Here we involved all measured data in the pH range between 3 and 9. We think that the quality of these values calculated out of much more measured data is higher than this of the old values.
We fitted additionally all 5 titration curves with the program
protofit [1]. It results in quite similar values in comparison to those from Hyperquad 2006 (see Table). This underlines the reliability of the new data. The discrepancy in the third pK_a is probably due to the mixture of amino and hydroxyl groups and is highly variable in dependence of the ending pH (varying between 9 and 10) which is also reflected in the large standard deviation.
We decided to fit only up to pH 9.3. (The protofit data are not involved in the manuscript; they should only support our Hyperquad 2006 values.)

	old (Hyperquad2000)	new (Hyperquad2006)	protofit
pK_{a1}	4.21 ± 0.07	4.55 ± 0.02	4.60 ± 0.03
pK_{a2}	6.01 ± 0.11	6.31 ± 0.01	6.25 ± 0.04
pK_{a3}	9.72 ± 0.20	9.56 ± 0.03	9.84 ± 0.17

Furthermore it could be demonstrated in the literature that different models and numbers of sites with the same computer program as well as with different computer programs applied to the same titration data results in differing protonation constants [2, 3]. Hence, one can only compare data determined with the same background, method and computer code.

References

1 B. F. Turner, J. B. Fein: Protofit: *A program for determining surface protonation constants from titration data.* Computers & Geosciences **32** (2006) 1344-1356.

2 D. M. Borrok, J. B. Fein: *The impact of ionic strength on the adsorption of protons, Pb, Cd, and Sr onto the surfaces of Gram negative bacteria: testing non-electrostatic, diffuse, and triple-layer models.* J. Colloid Interface Sci. **286** (2005) 110-126.

3 J. B. Fein, J.F. Boily, N. Yee, D. Gorman-Lewis, B. F. Turner: *Potentiometric titrations of Bacillus subtilis cells to low pH and a comparison of modelling approaches.* Geochim. Cosmochim. Acta **69** (2005) 1123-1132.