SUPPORTING INFORMATION

Title: Ion Interaction Models and Measurements of Eu^{3+} Complexation: HEDTA in Aqueous Solutions at 25 ^{o}C Containing 1:1 Na^{+} Salts and Citrate pH Buffer

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This document contains Table S1, and Figures S1 to S3, which are supporting information to the above publication. Table S1 lists the types of solutions measured by Schunk and Maurer (ref. 7 in the paper), and Figures S1 to S3 show model-calculated speciation in various citrate, HEDTA, and Eu^{3+}-containing solutions.

Table S1. Measurements of Osmotic Coefficients Made by Schunk and Maurer.[7]

Solution	**Compositions**	**Pitzer Model Interactions**	**Notes**
Na_3Citr	0.465 to 1.683 mol kg^{-1}	β^0, β^1, and C^ϕ for Na^+ - $Citr^{3-}$ interactions	
NaCl + H_3Citr	Up to 2.924 mol kg^{-1} NaCl, and 5.647 mol kg^{-1} H_3Citr	λ for NaCl - H_3Citr interactions	The acid dissociates very little in these solutions.
$NaNO_3$ + H_3Citr	Up to 2.906 mol kg^{-1} $NaNO_3$, and 3.861 mol kg^{-1} H_3Citr	λ for $NaNO_3$ - H_3Citr interactions	The acid dissociates very little in these solutions.
Na_3Citr + H_3Citr	Various ratios, up to 1.670 mol kg^{-1} Na_3Citr and 3.643 mol kg^{-1} H_3Citr	Three pairs of β^0, β^1, and C^ϕ parameters for Na^+-($Citr^{3-}$, $HCitr^{2-}$, H_2Citr^-) interactions, the self interaction of the aqueous acid ($\lambda_{n,n}$) plus many ternary (mixture) parameters	The compositions of the solutions were chosen so that all of the different species are present.

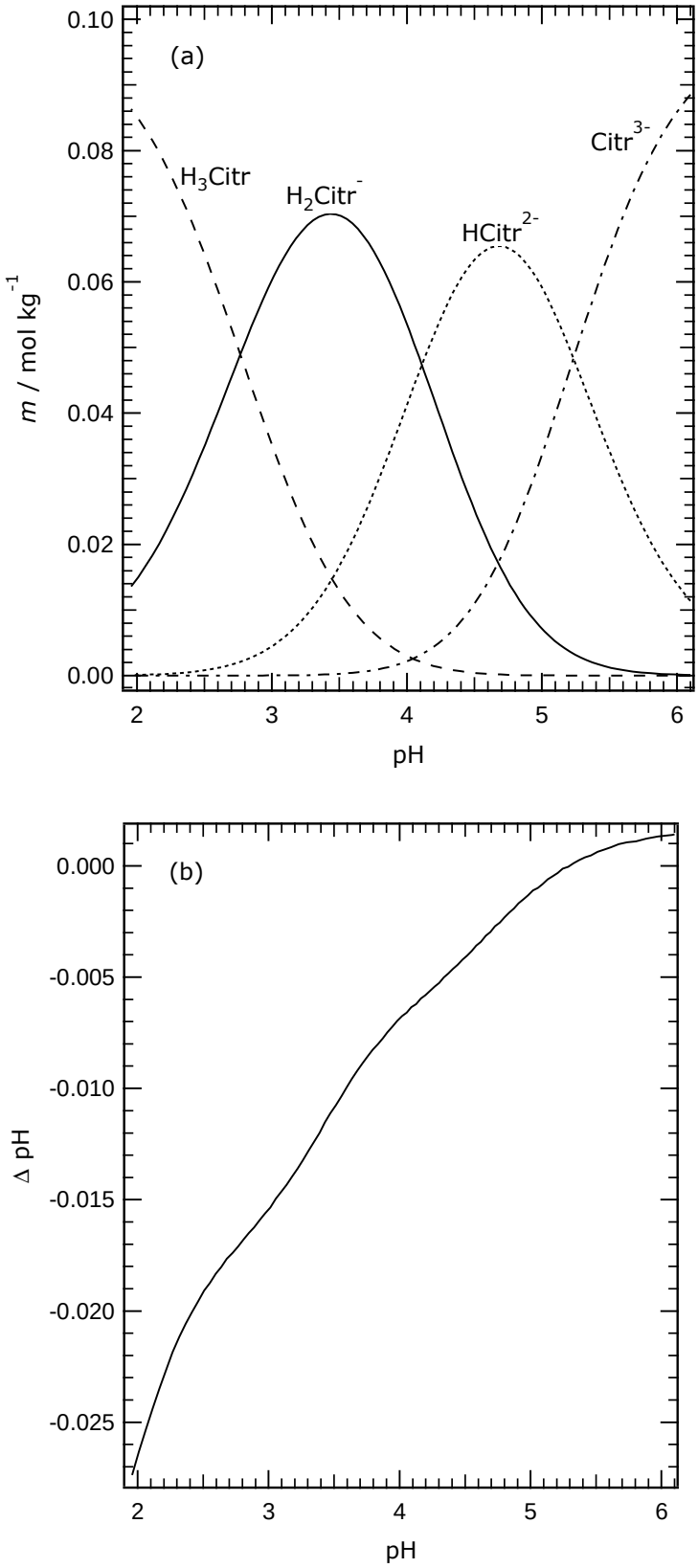

Figure S1. (a) Molalities (m) of citrate species in 1.0 mol kg^{-1} aqueous $NaNO_3$ at 25 °C, as a function of pH, calculated using the Pitzer model developed in this study. The solutions also contain 0.1 mol kg^{-1} total citrate, either as H_3Citr (at the lowest pH) or with substitution of some of the acid by Na_3Citr (at higher pH). (b) The effect on the calculated pH of a 1.0 mol kg^{-1} aqueous NaCl medium instead of aqueous $NaNO_3$. The quantity ΔpH is the difference between the calculated pH in aqueous NaCl and that in aqueous $NaNO_3$ (pH[NaCl] – pH[$NaNO_3$]), and it is plotted against the calculated pH in aqueous $NaNO_3$, for which the citrate speciation is shown in panel (a).

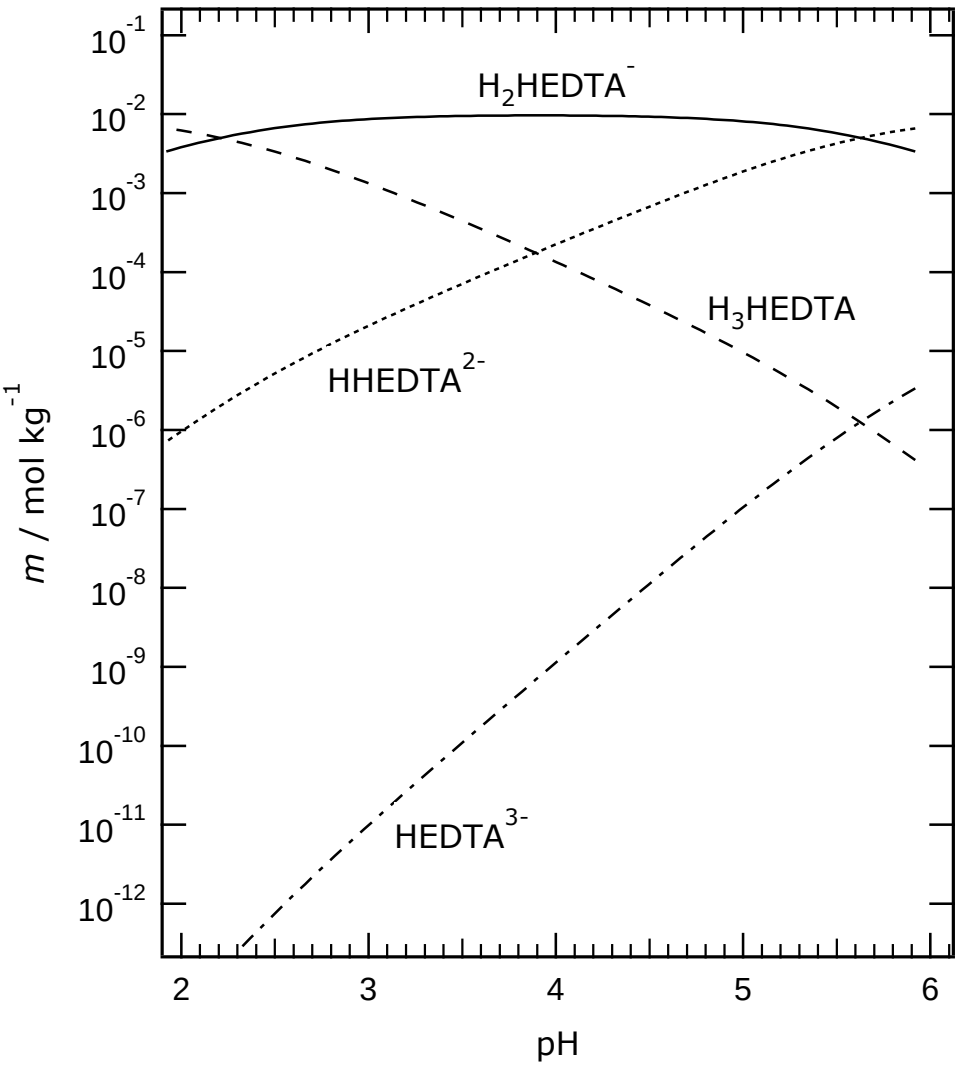

Figure S2. Molalities (*m*) of HEDTA species in 1.0 mol kg^{-1} aqueous $NaNO_3$ at 25 ^oC, as a function of pH, calculated using the Pitzer model developed in this study. The solutions also contain 0.1 mol kg^{-1} total citrate, either as H_3Citr (at the lowest pH) or with substitution of some of the acid by Na_3Citr (at higher pH), and 0.01 mol kg^{-1} of total HEDTA. The ionic strength of the solutions, calculated from the molalities of the ions, ranges from about 1.02 to 1.57 mol kg^{-1}.

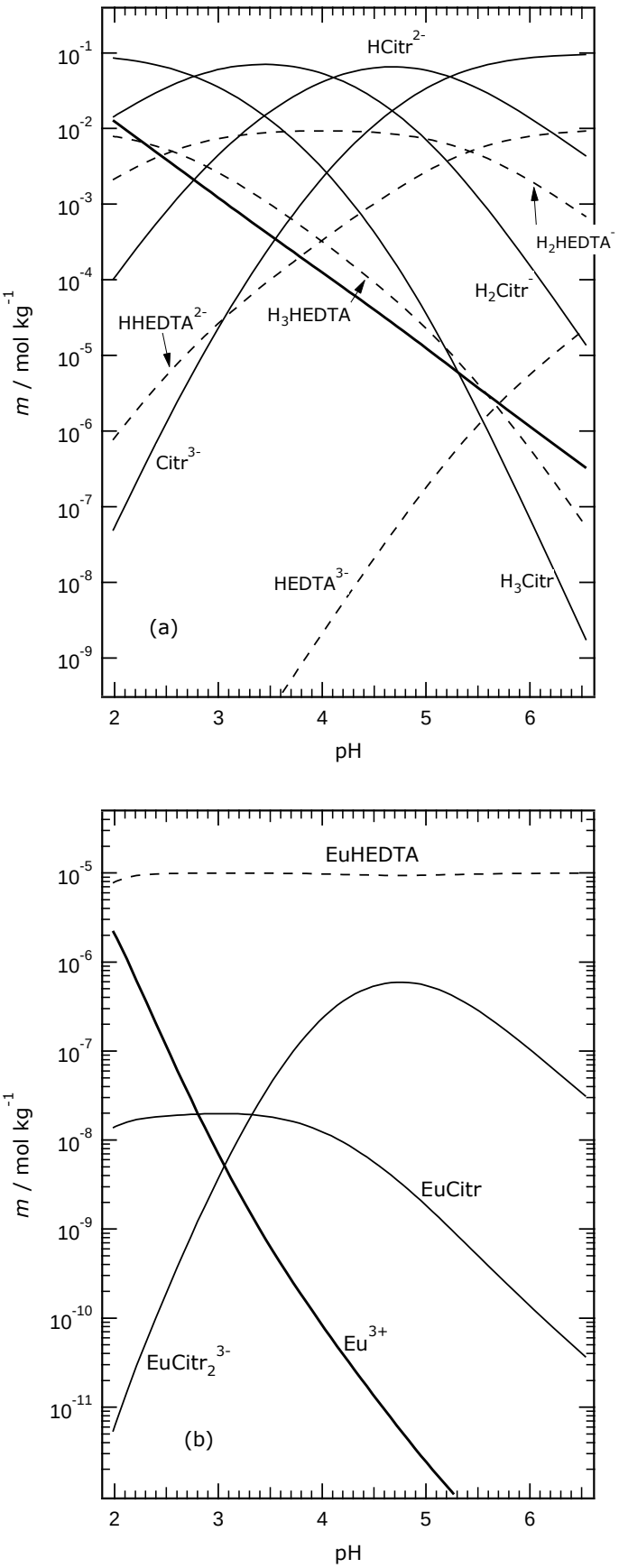

Figure S3. Calculated species molalities (*m*) in a solution containing 1.0 mol kg^{-1} $NaNO_3$, 0.10 mol kg^{-1} total citrate, 0.01 mol kg^{-1} total HEDTA, and 1.0×10^{-5} mol kg^{-1} Eu^{3+}, as a function of pH. (a) Acid – base equilibria (citrate and HEDTA speciation). Lines: solid (thick) – free H^+; solid (thin) – $Citr^{3-}$, $HCitr^{2-}$, H_2Citr^-, and H_3Citr as indicated; dashed – $HEDTA^{3-}$, $HHEDTA^{2-}$, H_2HEDTA^- and H_3HEDTA as indicated. (b) Europium speciation. Lines: solid (thick) – free Eu^{3+}; solid (thin) – $EuCitr_2^{3-}$ and EuCitr, as indicated; dashed – EuHEDTA.