Table S1. Impurities detected in the natural hematite sample as obtained by EMP analysis.

Element	Atomic %	Element	Atomic %
Na	0.006357	Cr	0.003167
Mn	0.002988	K	0.000106
Ti	0.004159	Cl	0.000765
Al	0.000382	Ni	0.001235
V	0.006886	Co	0.043010
P	0.002316	Cu	0.005196
Mg	0.004565	Au	0.004229

Single site/single protonation step TLM modeling

We initially tested our model using the formal charge as suggested by the 1-pK model (z=-1/2). Unfortunately, we did not obtain a satisfactory shape of the surface potential for any electrolyte affinities constant and capacities values (see Supporting Information). By varying the formal charge parameter (z), we have found that the experimental surface potential in the alkaline region can be relatively well reproduced by model assuming z=**-1** (as used in the first protonation step of the 2-pK model) (see Figure P2).

Reproducing the surface potential in an acidic pH region is much more problematic, and we were unable to provide the realistic model within the framework of the 1-pK model (z=-1/2) or the partial 2-pK model. However, for model with z=0 we observe the similar shape of the surface potential, but with a slightly increased magnitude.

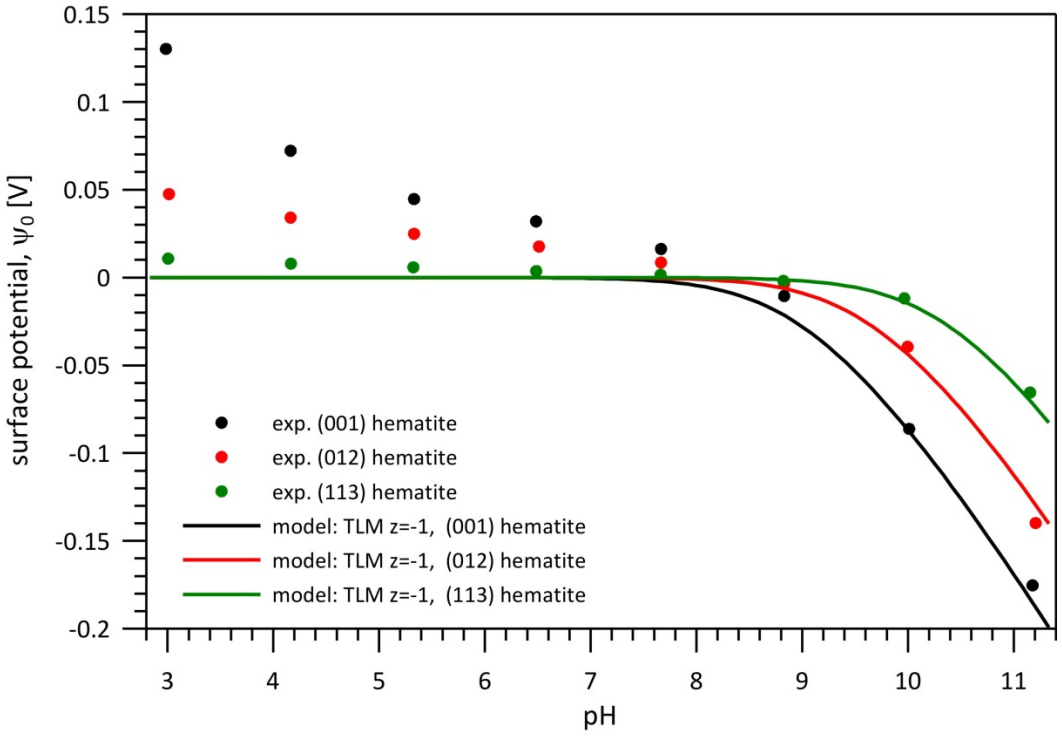

Figure S1: Fitting of the single-site MUSIC TLM model (z=0) for three considered hematite crystal faces. Model parameter values are collected in Table T1.

Table S2. Parameter values for the single-site/single-protonation step TLM model with z=0. Site densities for all considered crystal faces of hematite are estimated based on the site density of the perfectly cut hematite crystal, and are not treated as the best-fit parameters.

Parameter	hematite (001)	hematite (012)	hematite (113)
$\log K_H$	12.87	13.51	14.28
$\log K_{Na+}$	2.5	2.5	2.5
$\log K_{NO3-}$	2.5	2.5	2.5
c_x	0.42	0.42	0.42
c_2	0.66	0.66	0.66
N_s [sites/nm^2]	13.3	14.6	16.5

Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics