Correction to Force Field and a Surface Model Database for Silica to Simulate Interfacial Properties in Atomic Resolution

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A correction to Figures 8 and 9 in the original paper is presented (corrected Figures 8 and 9).[1] Significant corrections are required for Figure 8 while Figure 9 remains essentially the same.
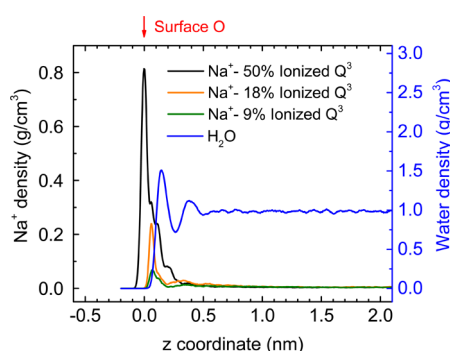

Figure 8. (corrected). Density profile of water and sodium ions on a regular Q^3 silica surface with different degree of ionization according to computation with the silica parameters embedded in the CHARMM force field (CHARMM-INTERFACE force field). The zero point of the z coordinate indicates the average position of silanol oxygen atoms on the surface (in the original figure, this position was erroneously shifted by 0.1 nm to the left). The density profile of water is essentially independent from the sodium content and shows the formation of two distinctive layers of decreasing intensity. The profile approaches a uniform density of 1.00 g/cm^3 at more than 0.5 nm away from the oxygen atoms of the surface. The distribution of sodium ions indicates major differences as a function of the degree of ionization. The penetration depth into the solution is highest for lowest ionization (see Figure 9 for details).

The original Figure 8 has two errors in axis labeling and was chosen from preliminary calculations with an earlier version of the published PCFF-INTERFACE force field.[2] The corrected Figure 8 uses corrected labels and calculations with the CHARMM-INTERFACE force field including the silica parameters as published.[1] The labeling errors in the original Figure 8 are the following. First, the density scale of the left y axis was erroneously plotted 1 order of magnitude lower in the original Figure 8; the sodium density can reach up to ∼0.8 g/cm^3 on highly ionized silica surfaces (corrected Figure 8) rather than 0.08 g/cm^3 as incorrectly suggested earlier (original Figure 8). Second, the average z coordinate of the surface oxygen atoms of silica was indicated to be at 0.0 nm in the original Figure 8; the real position was at $z = 0.1$ nm (an average of the positions of superficial Si atoms and O atoms was taken as a reference for $z = 0.0$ nm). The errors in axis labeling are fixed in the corrected Figure 8. The differences related to using an earlier version of the force field are as follows. The line shape of
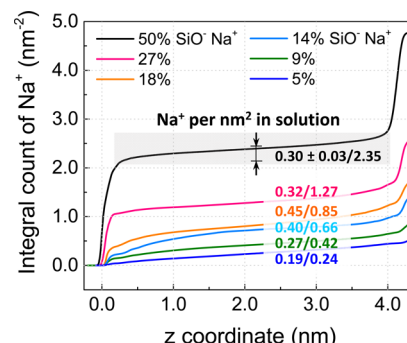

Figure 9. (corrected). Integral count of sodium ions per nm^2 between two aqueous Q^3 silica surfaces as a function of distance from superficial oxygen atoms of SiO(H, Na) groups for different percentage of silanol ionization according to atomistic simulation. The zero point of the z coordinate indicates the average position of silanol oxygen atoms on the left surface (in the original paper, this position was erroneously shifted by 0.1 nm to the left). The two numbers in each graph indicate the cumulative amount of dissociated sodium ions, defined as >0.3 nm away from surface oxygen atoms, in relation to the total available number of sodium ions per surface area. A higher total charge density first increases and then decreases sodium dissociation, with a maximum amount of dissociated sodium ions near 20% surface ionization (∼0.9 SiO$^-$ Na$^+$ per nm^2). The trend is consistent with zeta potentials, swelling observations on silica, and similar observations on clay minerals as a function of the area density of cations (see original text).

the ion density profiles with the CHARMM-INTERFACE force field in the corrected version is almost the same as with the preliminary version of the PCFF-INTERFACE force field in the original version. Some differences in the water density profiles are seen, however, which are associated with the use of the flexible SPC water model (as implemented in CHARMM) in the corrected Figure 8 and of the PCFF water model in the original Figure 8. The flexible SPC water model shows two distinguishable water layers on the silica surface while the PCFF water model shows three distinguishable water layers (shown for Q^3 silica surfaces with 0% ionization in both cases). This difference is related to the characteristics of the two different water models and should not be considered an error in the calculations.

The corrected Figure 9 can hardly be distinguished from the original Figure 9; it is only marginally affected by corrections in Figure 8. The data in the corrected Figure 9 are obtained by a uniform stretch by 0.1 nm, from $z = 0.0$ nm to $z = -0.1$ nm,

along the z coordinate from the original Figure 9. The integral count of dissociated sodium ions remains the same.

All other data and interpretations reported in the original paper are true and correct. The authors apologize for the inconvenience caused by human error.

Please note that the e-mail address of the corresponding author, Hendrik Heinz, has changed. The correct email address is hendrik.heinz@colorado.edu.

■ REFERENCES

(1) Emami, F. S.; Puddu, V.; Berry, R. J.; Varshney, V.; Patwardhan, S. V.; Perry, C. C.; Heinz, H. *Chem. Mater.* **2014**, *26*, 2647.

(2) Patwardhan, S. V.; Emami, F. S.; Berry, R. J.; Jones, S. E.; Naik, R. R.; Deschaume, O.; Heinz, H.; Perry, C. C. *J. Am. Chem. Soc.* **2012**, *134*, 6244.