Supplementary Information

Water and ions in electrified silica nano-pores: a molecular dynamics study

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Content:

1. Simulation Setup Details

Table S1 – Details of the different simulations done in the current study.

Run ID	Sim set	# of sims	Box Voltage	Gap Voltage	Ion conc. (M)	# silica slabs	Cations (#)	Anions (#)	Water (#)	Cumulat. time (ns)
256	1	3	0	0	0	2	0	0	8000	30
257	2	3	0	0	0.042	2	4	4	8000	30
258	3	3	0	0	0.1	2	12	12	8000	30
259	4	3	0	0	0.21	2	24	24	8000	30
260	5	3	0	0	0.42	2	50	50	8000	30
261	6	3	0	0	0.84	2	100	100	8000	30
262	7	3	0	0	2	2	238	238	8000	30
214	8	6	0.2	0.08	0	2	0	0	8000	60
230	9	6	0.2	0.08	0.021	2	2	2	8000	60
213	10	6	0.2	0.08	0.042	2	4	4	8000	60
236	11	6	0.2	0.08	0.0565	2	6	6	8000	60
212	12	6	0.2	0.08	0.1	2	12	12	8000	60
211	13	6	0.2	0.08	0.21	2	24	24	8000	60
210	14	6	0.2	0.08	0.42	2	50	50	8000	60
209	15	6	0.2	0.08	0.84	2	100	100	8000	60
221	16	6	0.2	0.08	2	2	238	238	8000	60
220	17	6	0.5	0.2	0	2	0	0	8000	60
231	18	6	0.5	0.2	0.021	2	2	2	8000	60
219	19	6	0.5	0.2	0.042	2	4	4	8000	60
237	20	6	0.5	0.2	0.0565	2	6	6	8000	60
218	21	6	0.5	0.2	0.1	2	12	12	8000	60
217	22	6	0.5	0.2	0.21	2	24	24	8000	60
216	23	6	0.5	0.2	0.42	2	50	50	8000	60
215	24	6	0.5	0.2	0.84	2	100	100	8000	60
186	25	6	1	0.4	0	2	0	0	8000	60
227	26	6	1	0.4	0.021	2	2	2	8000	60
191	27	6	1	0.4	0.042	2	4	4	8000	60
232	28	6	1	0.4	0.0565	2	6	6	8000	60
190	29	6	1	0.4	0.1	2	12	12	8000	60
189	30	6	1	0.4	0.21	2	24	24	8000	60
188	31	6	1	0.4	0.42	2	50	50	8000	60
182	32	6	1	0.4	0.84	2	100	100	8000	60
223	33	6	1	0.4	2	2	238	238	8000	60
201	34	6	5	2	0	2	0	0	8000	60

238	35	6	5	2	0.021	2	2	2	8000	60
200	36	6	5	2	0.042	2	4	4	8000	60
233	37	6	5	2	0.0565	2	6	6	8000	60
243	38	6	5	2	0.1	2	12	12	8000	60
242	39	6	5	2	0.21	2	24	24	8000	60
241	40	6	5	2	0.42	2	50	50	8000	60
240	41	6	5	2	0.84	2	100	100	8000	60
224	42	6	5	2	2	2	238	238	8000	60
197	43	6	10	4	0	2	0	0	8000	90
228	44	6	10	4	0.021	2	2	2	8000	90
196	45	6	10	4	0.042	2	4	4	8000	90
234	46	6	10	4	0.0565	2	6	6	8000	90
195	47	6	10	4	0.1	2	12	12	8000	90
194	48	6	10	4	0.21	2	24	24	8000	90
193	49	6	10	4	0.42	2	50	50	8000	90
192	50	6	10	4	0.84	2	100	100	8000	90
225	51	6	10	4	2	2	238	238	8000	90
207	52	6	20	8	0	2	0	0	8000	120
229	53	6	20	8	0.021	2	2	2	8000	120
206	54	6	20	8	0.042	2	4	4	8000	120
235	55	6	20	8	0.0565	2	6	6	8000	120
205	56	6	20	8	0.1	2	12	12	8000	120
204	57	6	20	8	0.21	2	24	24	8000	120
203	58	6	20	8	0.42	2	50	50	8000	120
202	59	6	20	8	0.84	2	100	100	8000	120
226	60	6	20	8	2	2	238	238	8000	120
85	61	6	0	0	0	0	0	0	2000	60
111	62	6	0	0	0.5	0	17	17	2000	60
109	63	6	0	0	1	0	35	35	2000	60
112	64	6	0	0	1.5	0	52	52	2000	60
128	65	6	0	0	2	0	68	68	2000	60
129	66	6	0	0	2.5	0	85	85	2000	60
										4.56 µs

2. Convergence of the Dielectric Constant

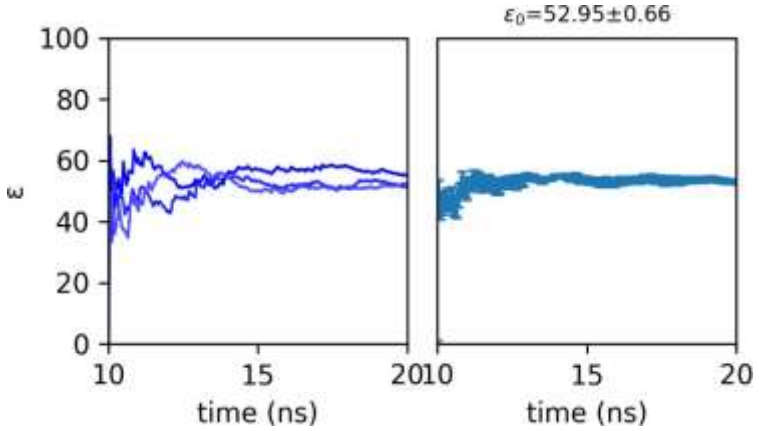

Figure S1 – The convergence of the dielectric constant for the system under an electric field of 8V and an ionic concentration of 2M. (a) shows the results for three individual simulations and (b) presents the average, with the error represented as bars gradually decreasing as the simulations converge.

3. Distribution of the water dipole in the x-direction of the surface

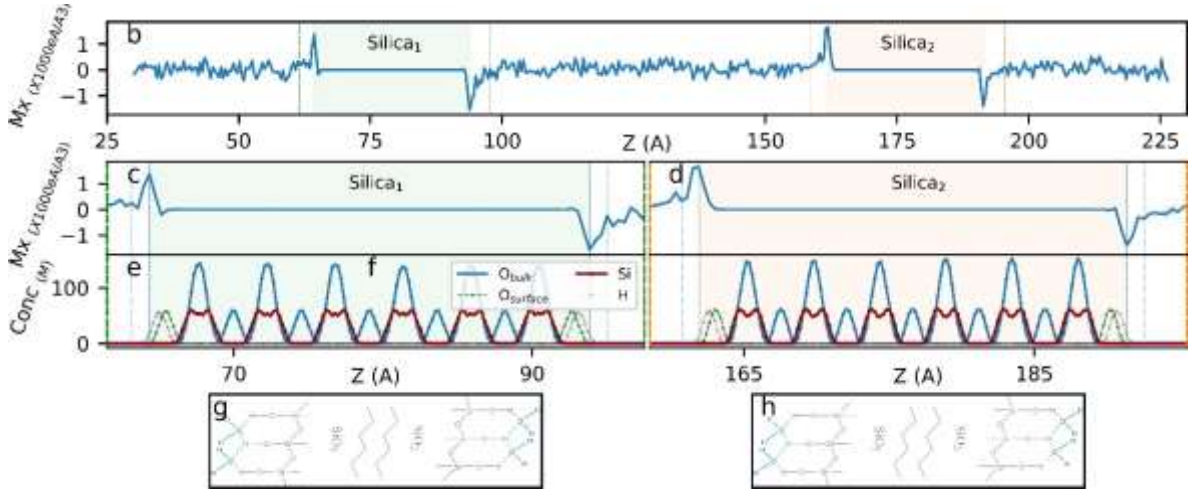

Figure S2 - The distribution of the x-component (M_x) of water molecules between two uncharged silica surfaces highlights the importance of hydrogen-bonds in orientation of water molecules. (a) M_x distribution along the z direction. Magnified view of M_x at the interface with (c) silica$_1$ (green highlighted in panel a) and (d) silica$_2$ (orange highlighted in panel) and various components of (e) silica$_1$ and (f) silica$_2$ illustrating (g) and (h) the water molecule dipoles orienting outwards from the surface. Errors are shown as shades around plotted values.

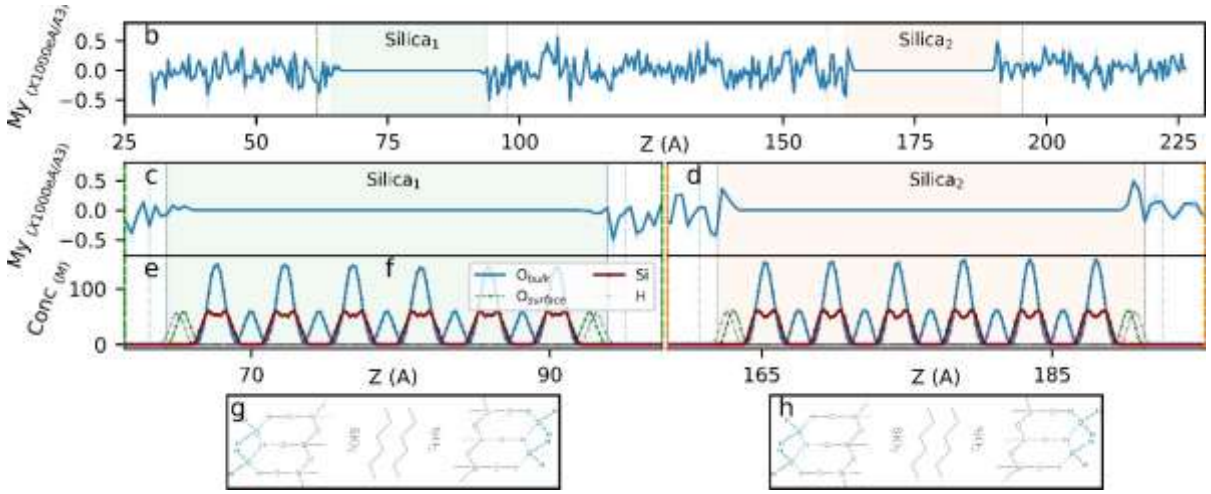

Figure S3 - The distribution of the y-component (M_y) of water molecules between two uncharged silica surfaces shows no effect of hydrogen bonding in the molecular organization in y direction. (a) M_x distribution along the z direction. Magnified view of M_x at the interface with (c) silica$_1$ (green highlighted in panel a) and (d) silica$_2$ (orange highlighted in panel) and various components of (e) silica$_1$ and (f) silica$_2$ illustrating no preferred water orientation near the silica surface in the y direction. g) and h) arrangement of water molecules near the silica surfaces.

4. Water dipole and ionic distributions in the gap at different applied voltages

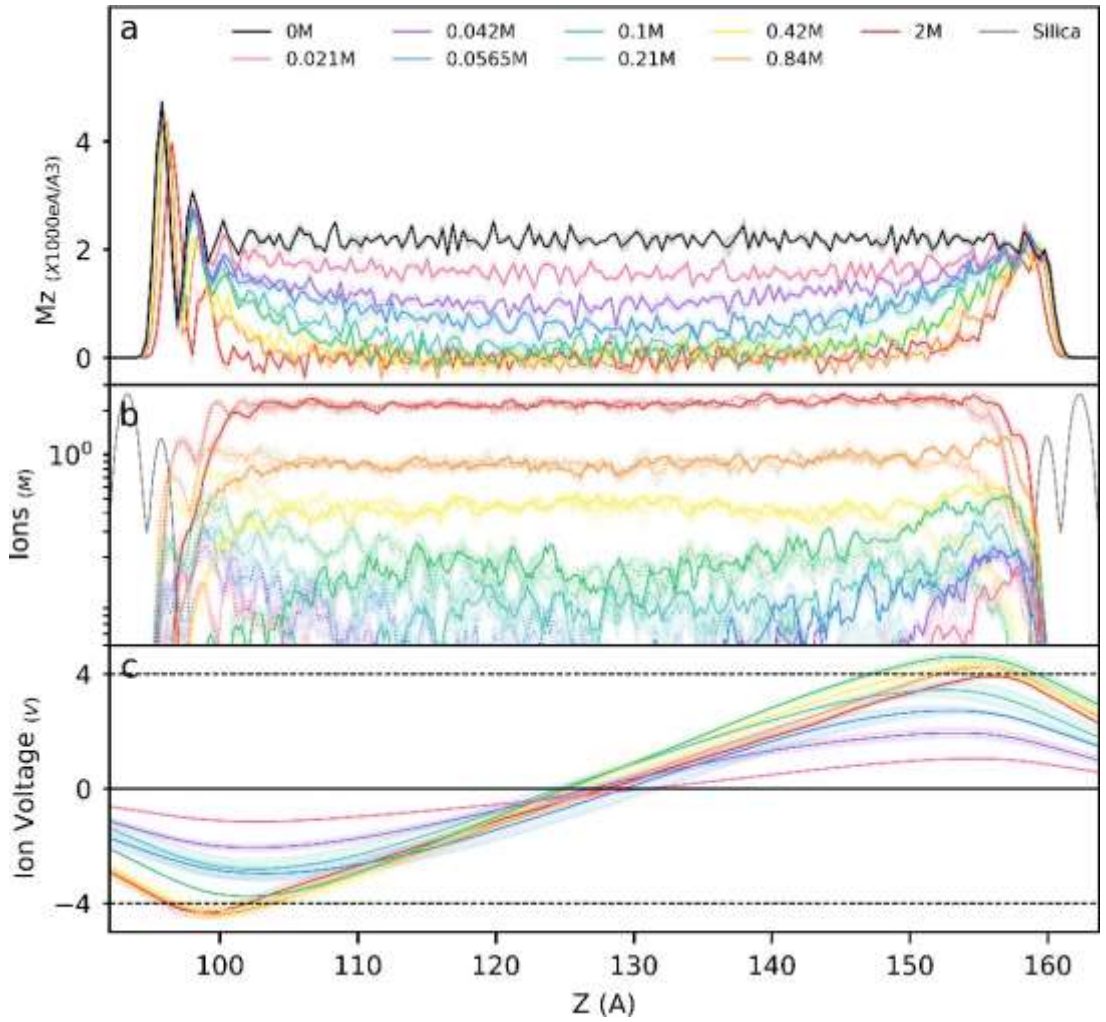

Figure S4 - The water dipole and ions distribution across the box, together with the amount of voltage screened under the externally applied 8 V. (a) The z-component of the water dipole (M_z), (b) local cation (solid line) and anion (dotted lines) concentrations and (c) ion-induced voltage distribution along the coordinate perpendicular to the silica plates (z). Errors are depicted as shades around the plots.

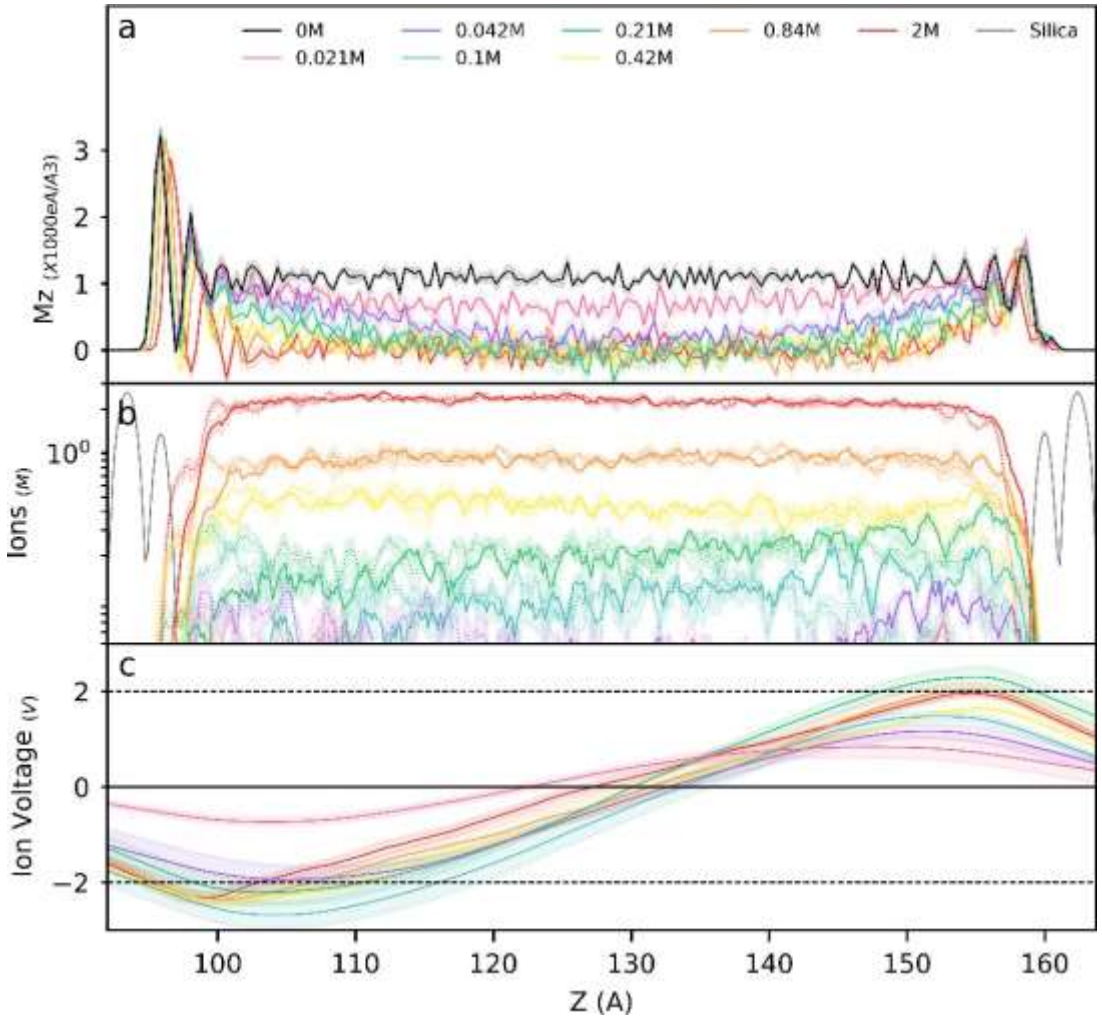

Figure S5 - The water dipole and ions distribution across the box, together with the amount of voltage screened under the externally applied 4 V. (a) The z-component of the water dipole (M_z), (b) local cation (solid line) and anion (dotted lines) concentrations and (c) ion-induced voltage distribution along the coordinate perpendicular to the silica plates (z). Errors are depicted as shades around the plots.

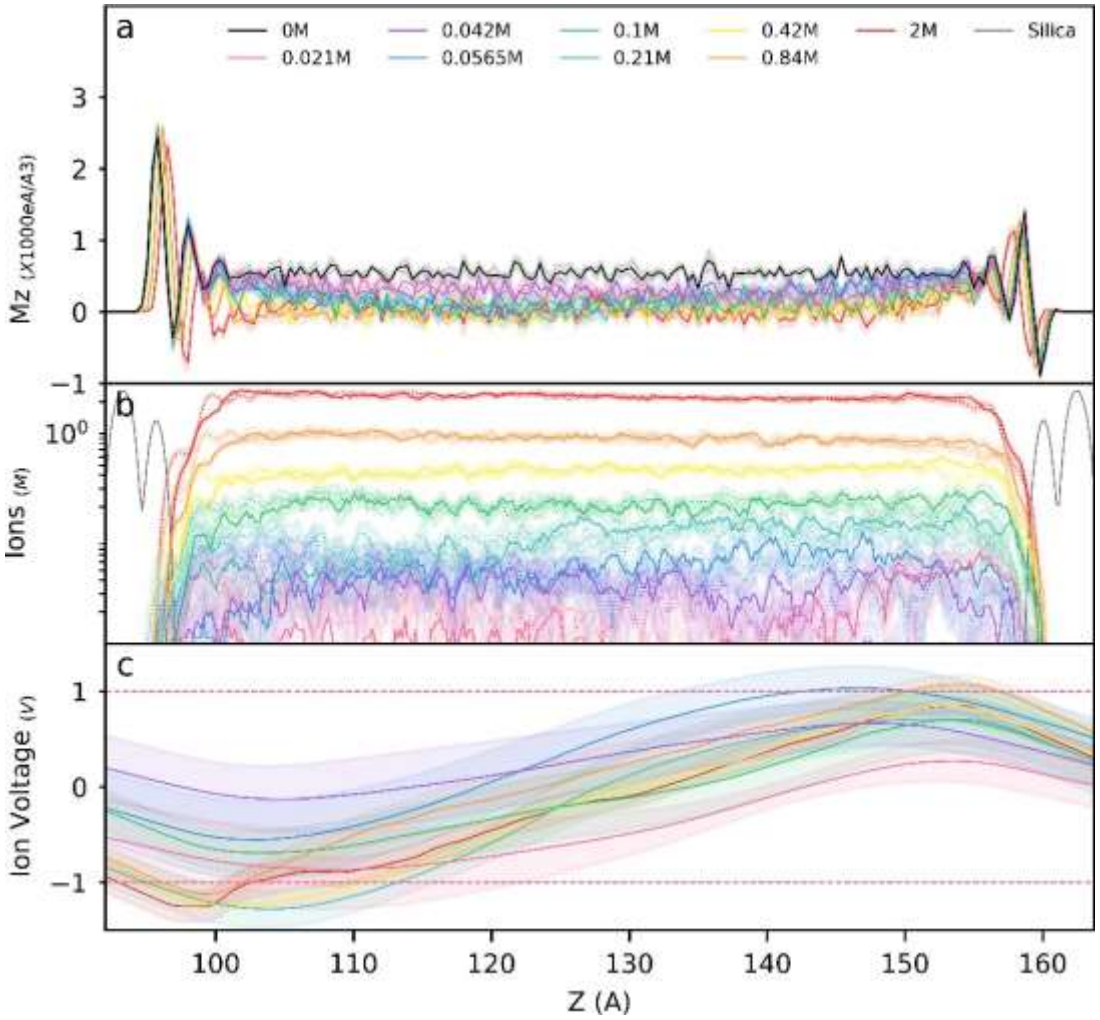

Figure S6 - The water dipole and ions distribution across the box, together with the amount of voltage screened under the externally applied 2 V. (a) The z-component of the water dipole (M_z), (b) local cation (solid line) and anion (dotted lines) concentrations and (c) ion-induced voltage distribution along the coordinate perpendicular to the silica plates (z). Errors are depicted as shades around the plots.

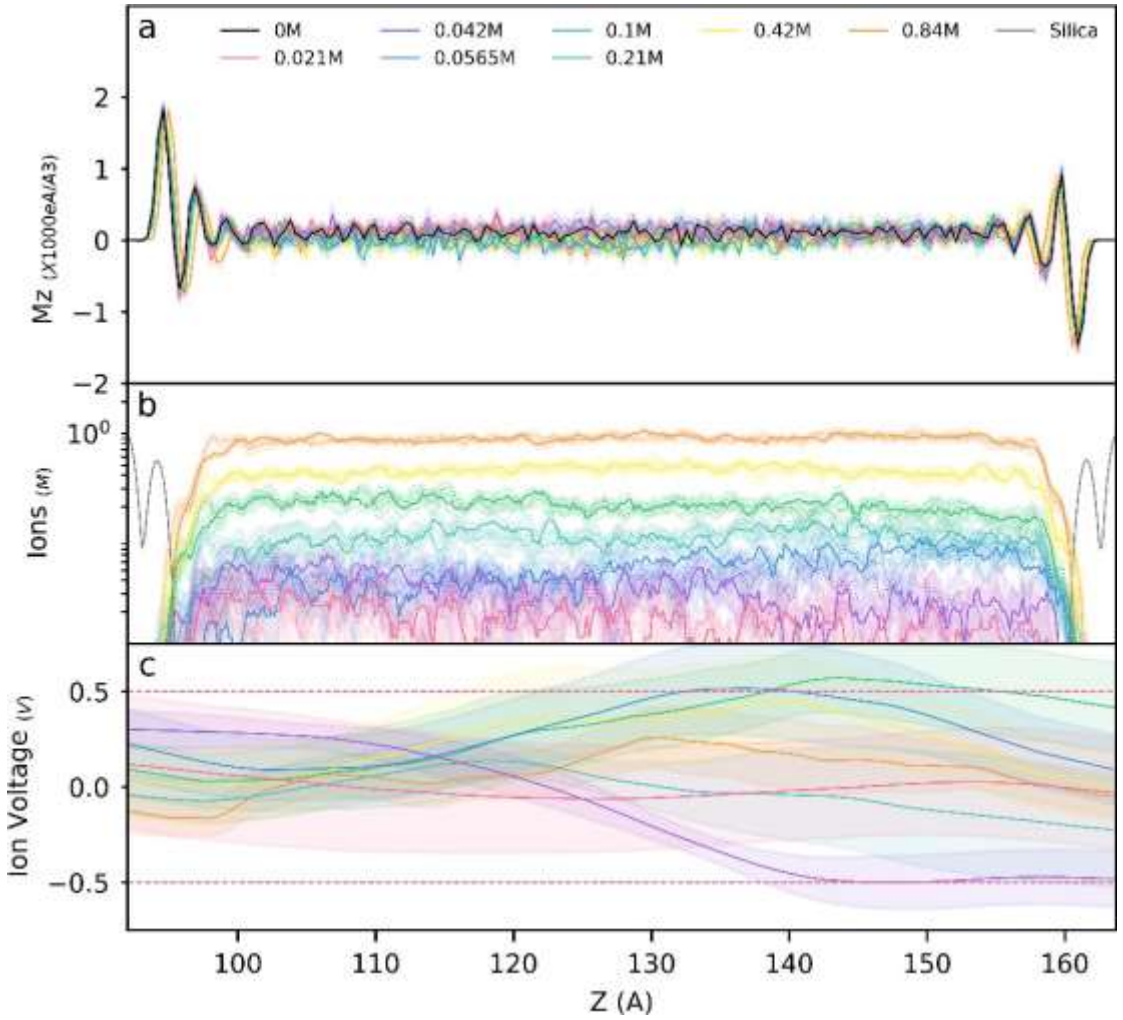

Figure S7 - The water dipole and ions distribution across the box, together with the amount of voltage screened under the externally applied 0.4 V. (a) The z-component of the water dipole (M_z), (b) local cation (solid line) and anion (dotted lines) concentrations and (c) ion-induced voltage distribution along the coordinate perpendicular to the silica plates (z). Errors are depicted as shades around the plots.

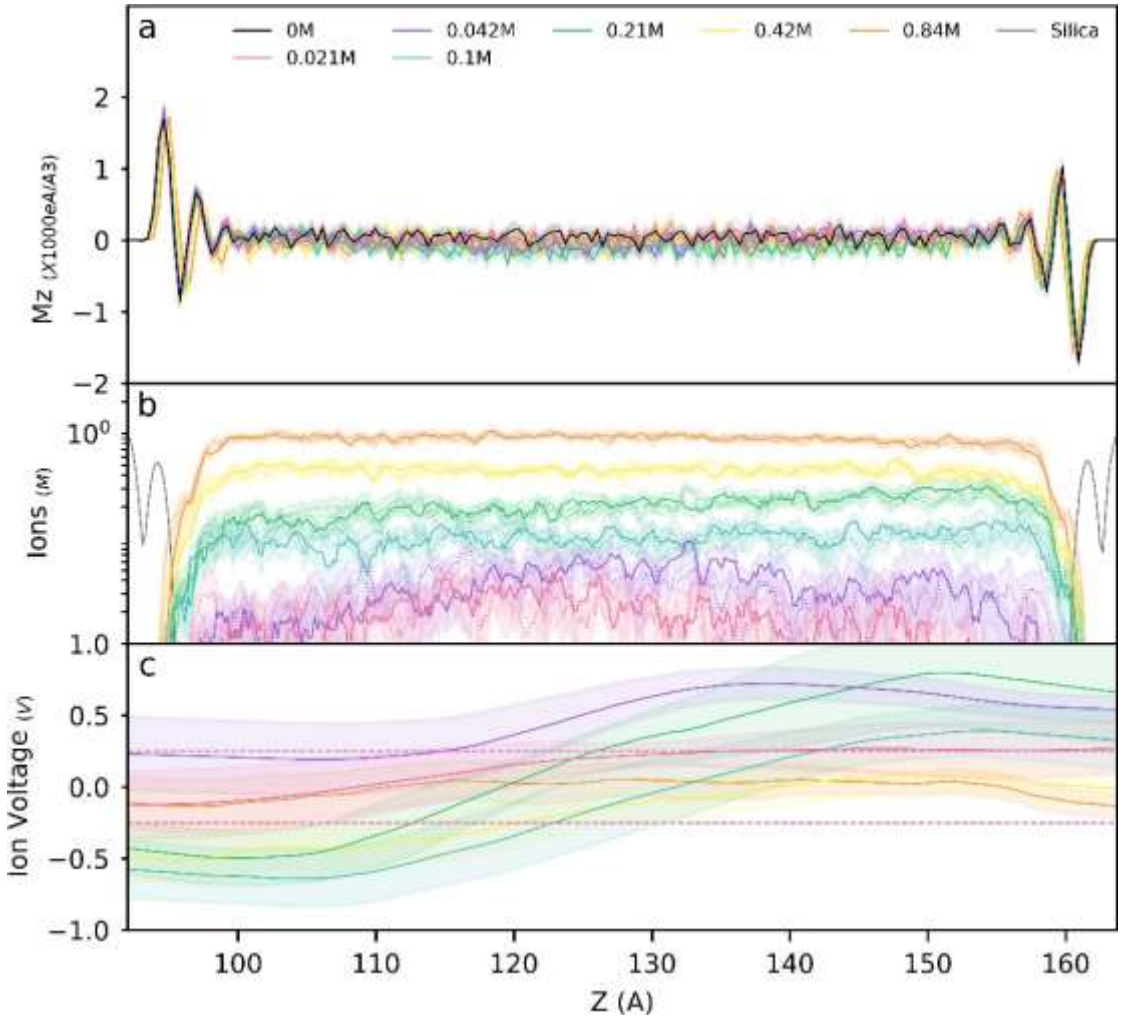

Figure S8 - The water dipole and ions distribution across the box, together with the amount of voltage screened under the externally applied 0.2 V. (a) The z-component of the water dipole (M_z), (b) local cation (solid line) and anion (dotted lines) concentrations and (c) ion-induced voltage distribution along the coordinate perpendicular to the silica plates (z). Errors are depicted as shades around the plots.

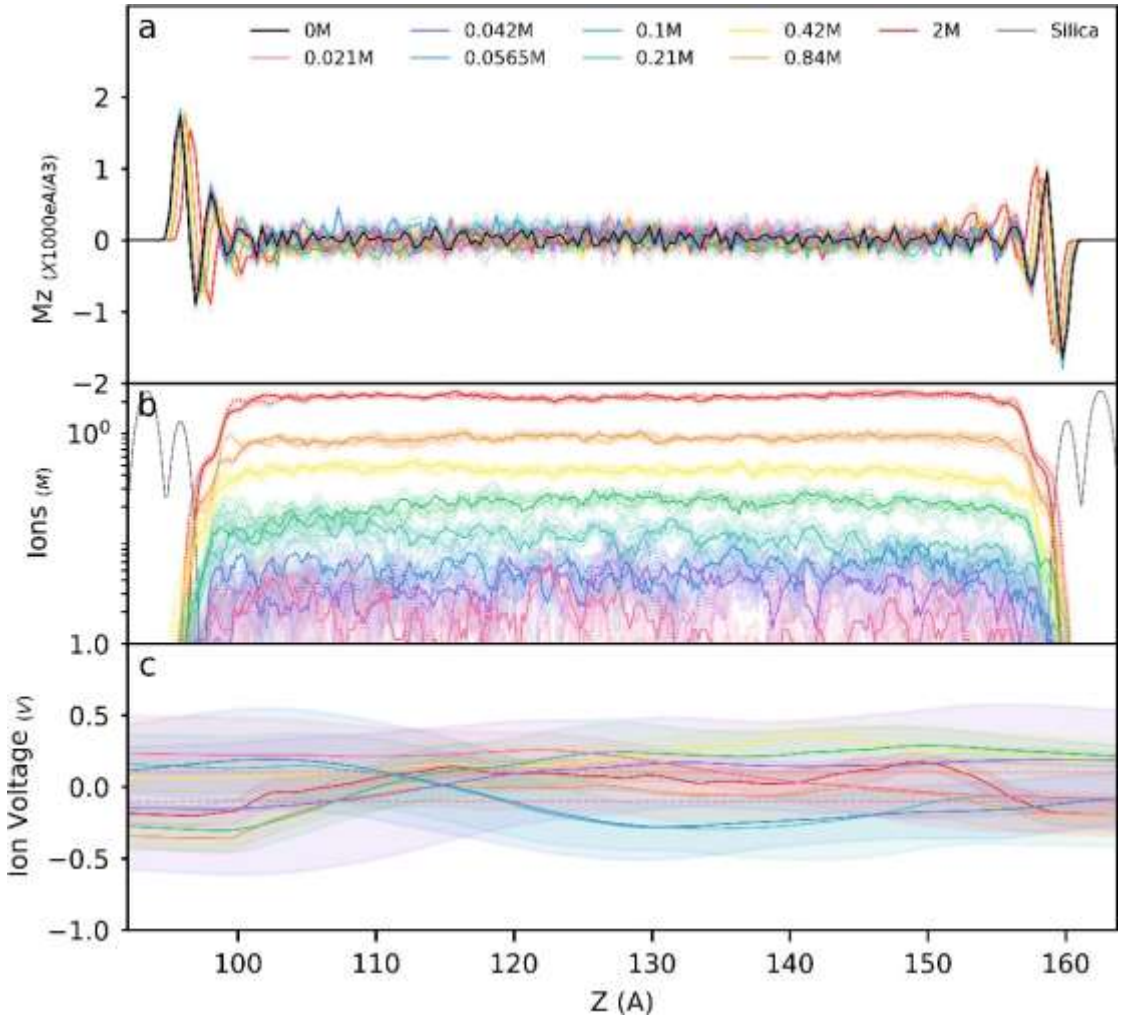

Figure S9 - The water dipole and ions distribution across the box, together with the amount of voltage screened under the externally applied 0.08 V. (a) The z-component of the water dipole (M_z), (b) local cation (solid line) and anion (dotted lines) concentrations and (c) ion-induced voltage distribution along the coordinate perpendicular to the silica plates (z). Errors are depicted as shades around the plots.

5. Water dipole and ionic distributions in the gap at different ionic concentrations

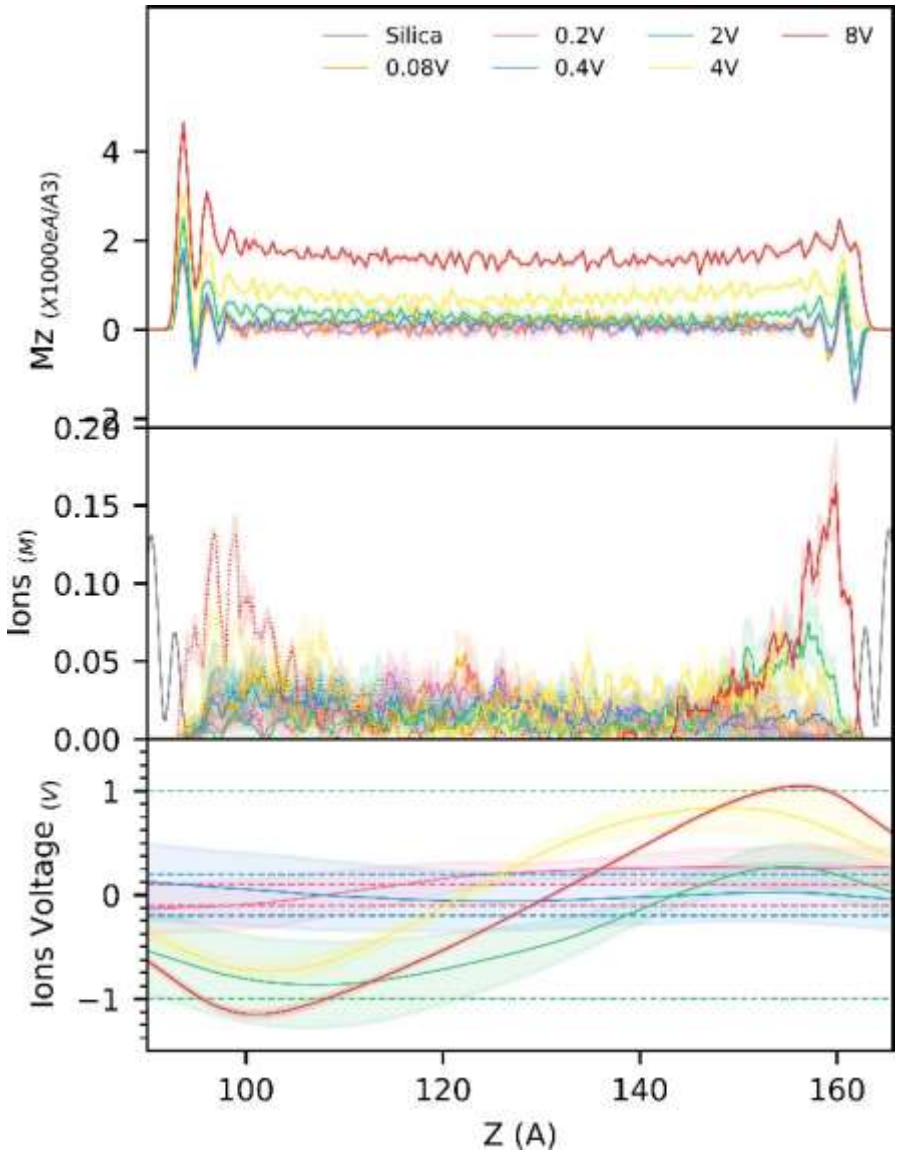

Figure S10 - The water dipole and ions distribution across the box, together with the amount of voltage screened by ions for ionic concentration of 0.021 M.(a) The z-component of the water dipole (M_z), (b) local cation (solid lines) and anion (dotted lines) concentrations and (c) ions voltage distribution along the coordinate perpendicular to the silica plates (z). Errors are depicted as shades around the plots.

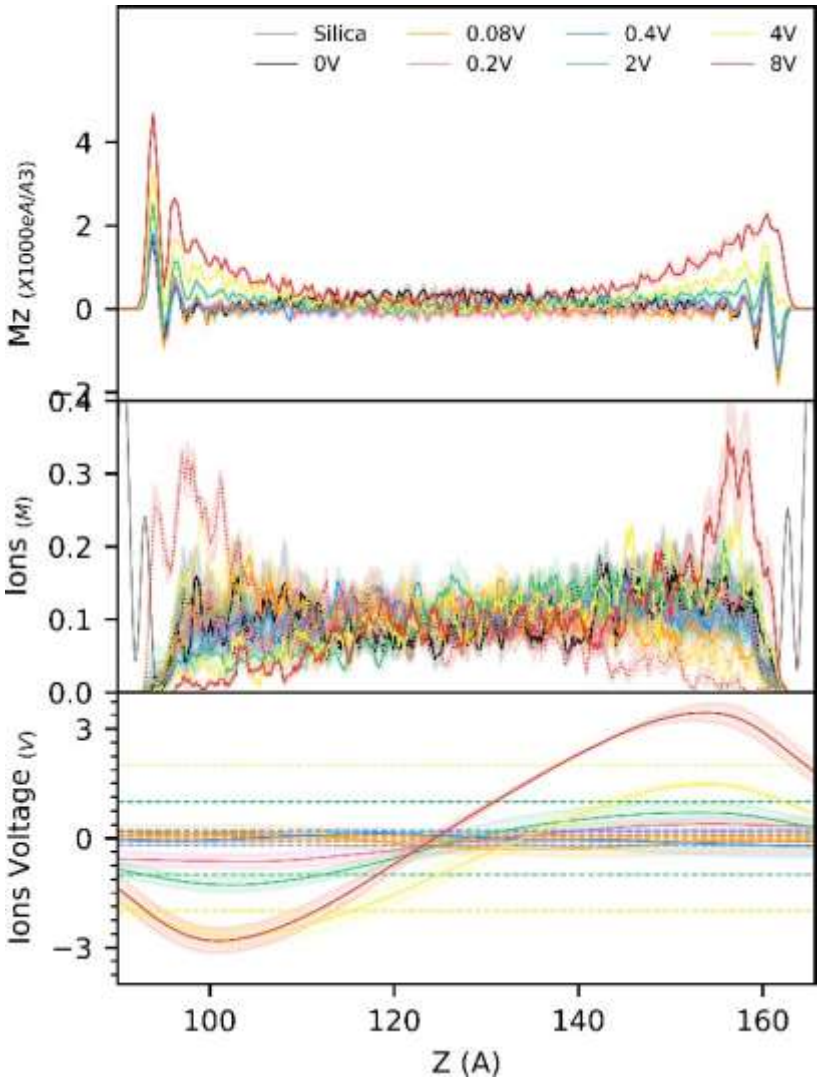

Figure S11 - The water dipole and ions distribution across the box, together with the amount of voltage screened by ions for ionic concentration of 0.042M.(a) The z-component of the water dipole (M_z), (b) local cation (solid lines) and anion (dotted lines) concentrations and (c) ions voltage distribution along the coordinate perpendicular to the silica plates (z). Errors are depicted as shades around the plots.

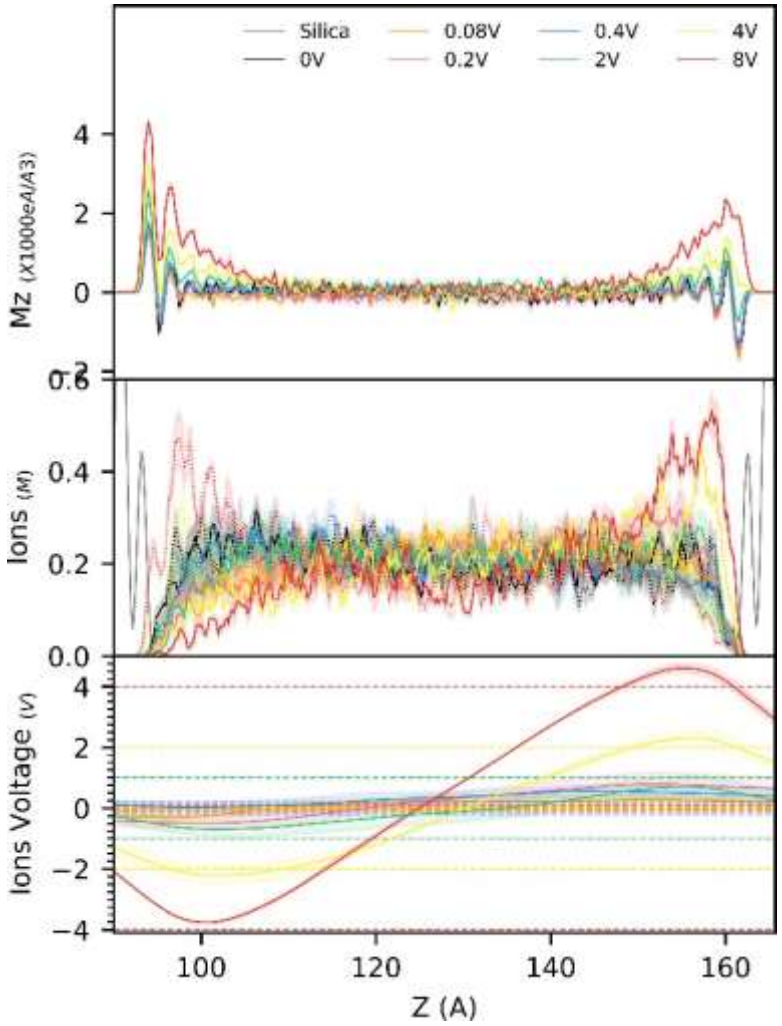

Figure S12 - The water dipole and ions distribution across the box, together with the amount of voltage screened by ions for ionic concentration of 0.21 M.(a) The z-component of the water dipole (M_z), (b) local cation (solid lines) and anion (dotted lines) concentrations and (c) ions voltage distribution along the coordinate perpendicular to the silica plates (z). Errors are depicted as shades around the plots.

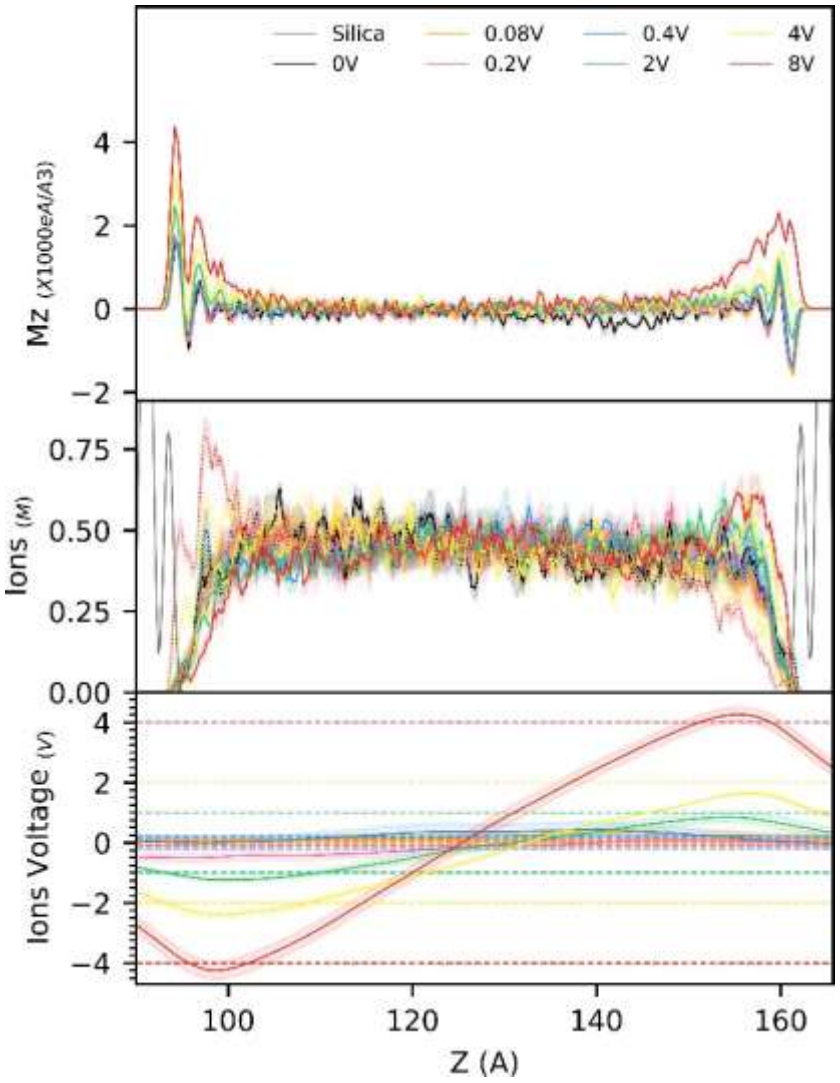

Figure S13 - The water dipole and ions distribution across the box, together with the amount of voltage screened by ions for ionic concentration of 0.42 M.(a) The z-component of the water dipole (M_z), (b) local cation (solid lines) and anion (dotted lines) concentrations and (c) ions voltage distribution along the coordinate perpendicular to the silica plates (z). Errors are depicted as shades around the plots.

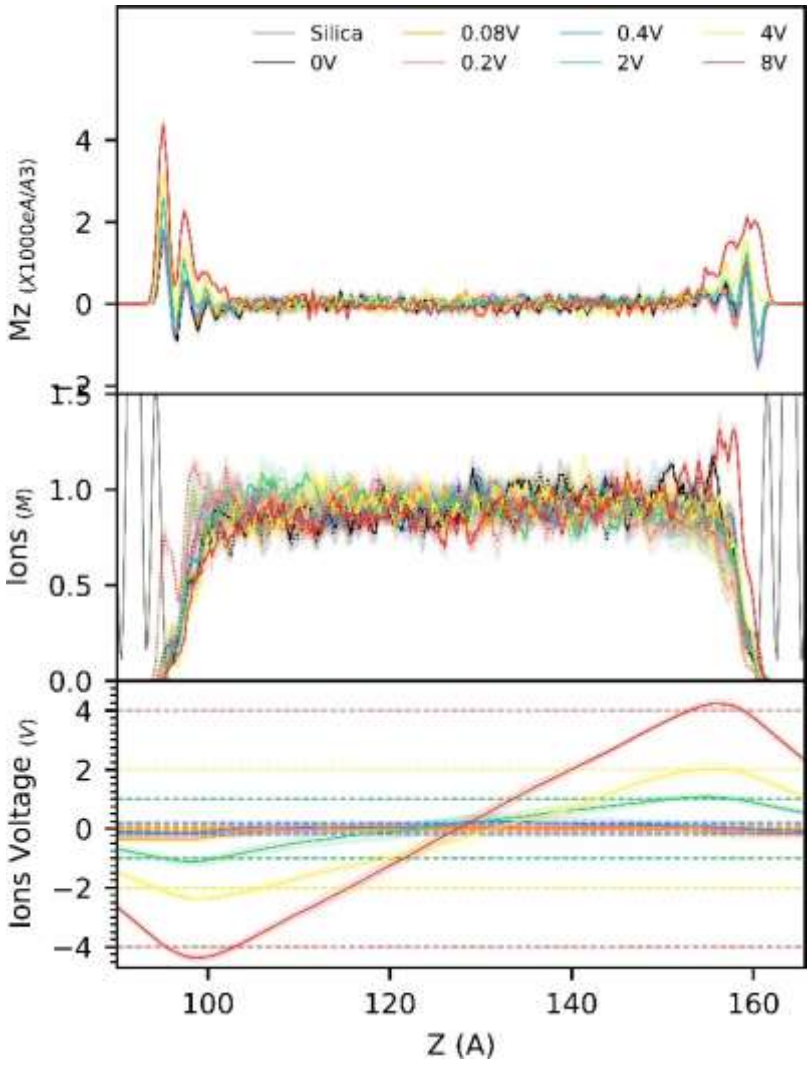

Figure S14 - The water dipole and ions distribution across the box, together with the amount of voltage screened by ions for ionic concentration of 0.84 M.(a) The z-component of the water dipole (M_z), (b) local cation (solid lines) and anion (dotted lines) concentrations and (c) ions voltage distribution along the coordinate perpendicular to the silica plates (z). Errors are depicted as shades around the plots.

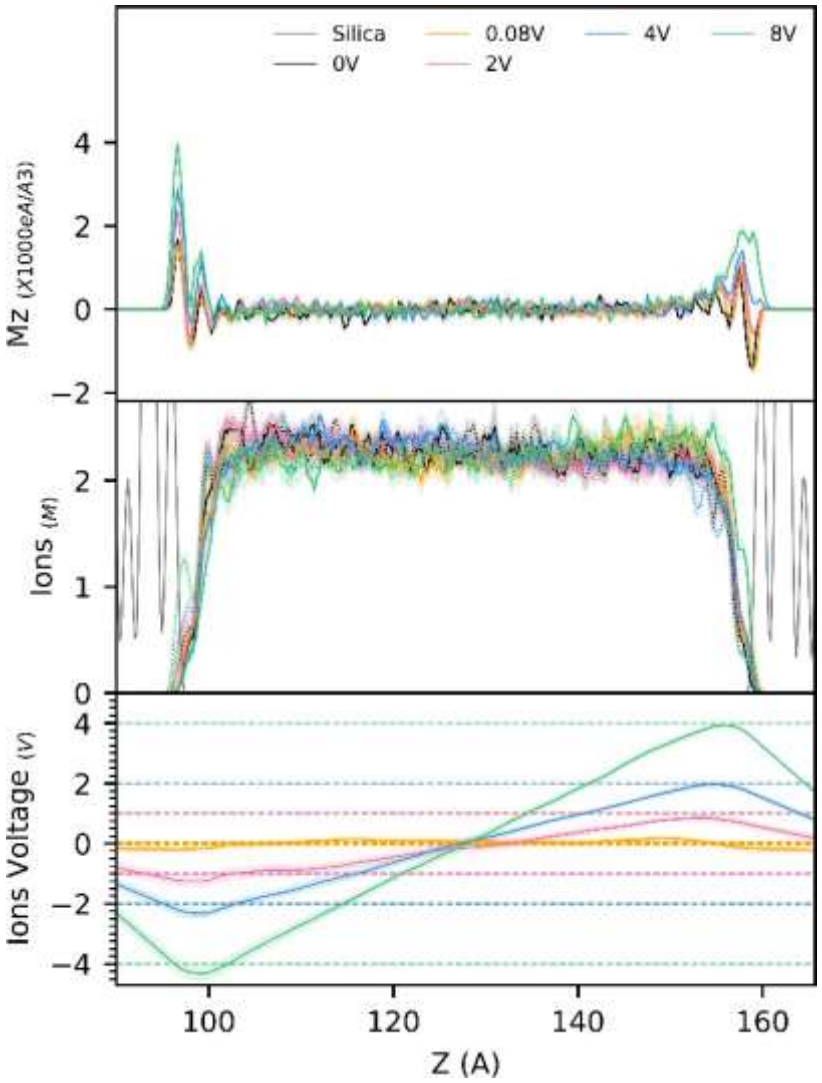

Figure S15 - The water dipole and ions distribution across the box, together with the amount of voltage screened by ions for ionic concentration of 2 M.(a) The z-component of the water dipole (M_z), (b) local cation (solid lines) and anion (dotted lines) concentrations and (c) ions voltage distribution along the coordinate perpendicular to the silica plates (z). Errors are depicted as shades around the plots.

6. Lateral organisation of water molecules at the surface of silica

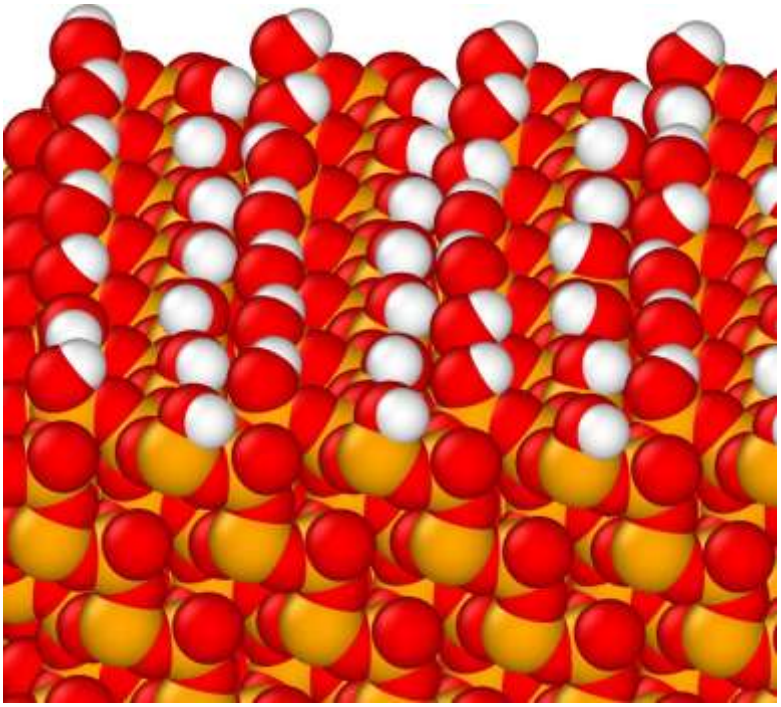

Figure S16 – Structure of the silica surface considered in the current study. Oxygen, Phosphate and Hydrogen atoms are coloured in red, orange and while respectively.