

# Errata for From Silica Nano-Particles to Silica Gels and Beyond

By Christian Sögaard

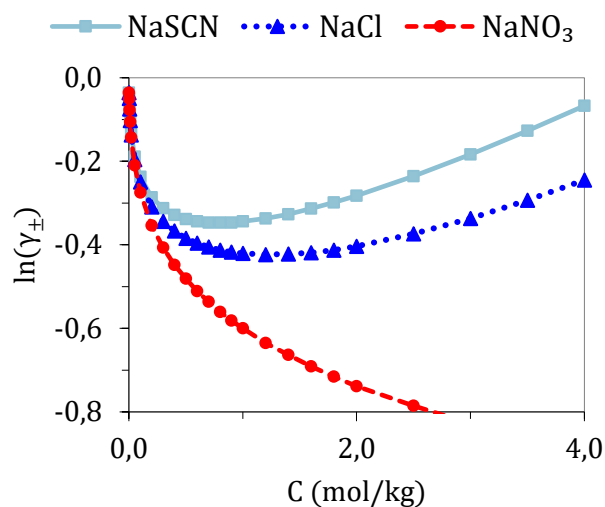
Page 11, line 18: “However, the rate of force increase is not the same.” Here force should say potential.

Page 40, last sentence: “K<sup>+</sup> and Ca<sup>2+</sup> as well as the large divalent ions...” Here Ca<sup>2+</sup> should say Cs<sup>+</sup>.

Page 42, line 5: “...not lead to 20% decrease of Na<sup>+</sup> concentration at the interface.” Here Na<sup>+</sup> should say Li<sup>+</sup>.

Supporting info for the articles is missing and can be found in order below:

Supporting info article II:



**Figure S1:** The natural logarithm of the mean ion activity coefficients of NaSCN, NaCl, and NaNO<sub>3</sub> as a function of concentration (moles of solute per kg of water). The values were obtained from experimental data by Hamer & Wu [1].

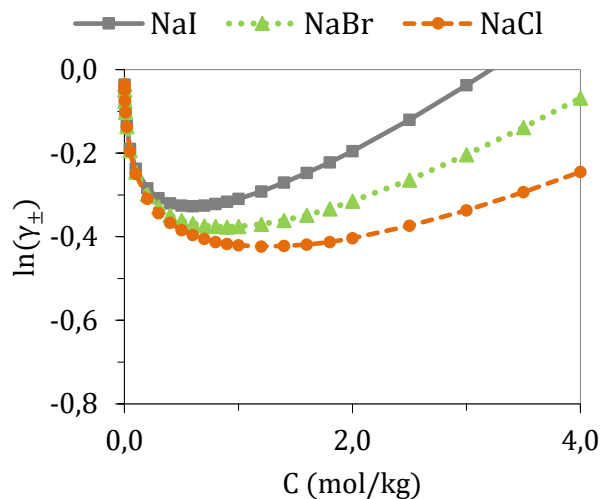


Figure S2: The natural logarithm of the mean ion activity coefficients of NaI, NaBr, and NaCl as a function of concentration (moles of solute per kg of water). The values were obtained from experimental data by Hamer & Wu [1].

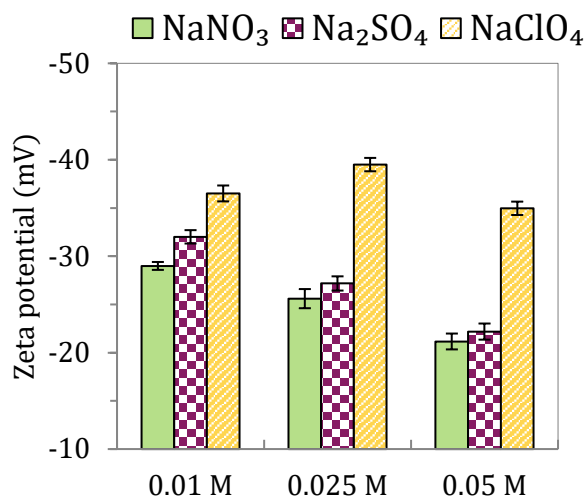
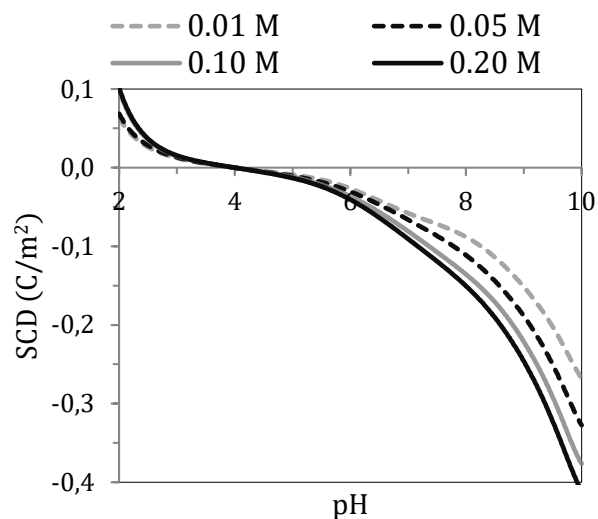
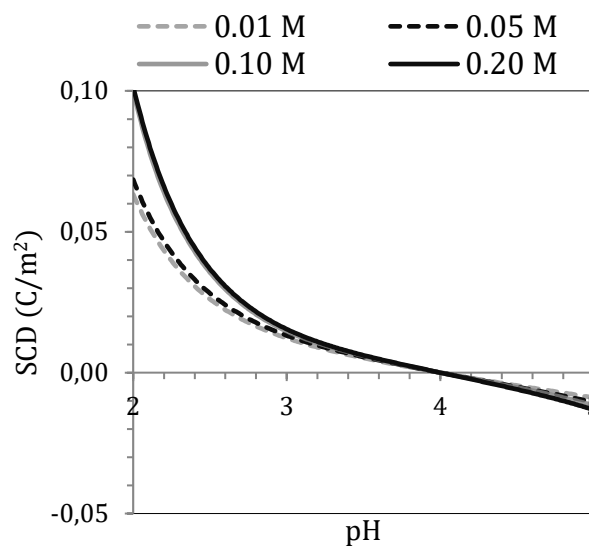


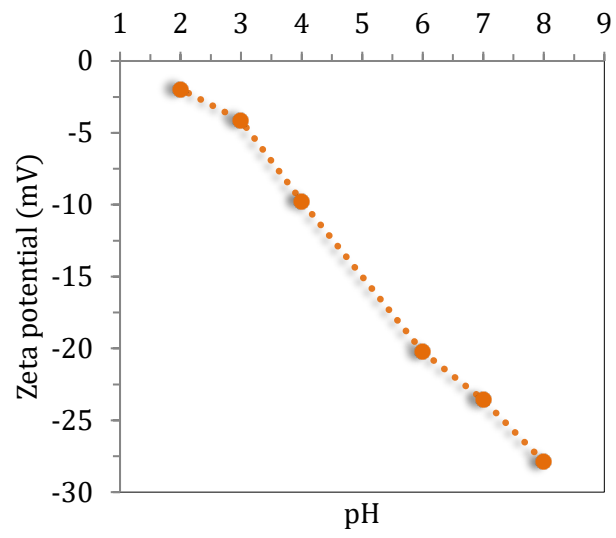
Figure S3. Column chart showing results from the zeta potential measurements of 2 wt% TM silica nanoparticles at varying cation concentrations and at pH 9.4. Twelve runs were performed for each cation concentration and the error bars represent the 95% confidence interval calculated from the standard deviations obtained from these.



**Figure S4.** Titration curves of 2 wt% TM silica nanoparticles in the presence of 0.20, 0.10, 0.05, and 0.01 M NaCl showing surface charge density as a function of pH.



**Figure S5.** Zoom in of the pH range 2-5 of the titration curves presented in Figure S4.



**Figure S6. Zeta potential measurements at pH 2.0-8.0 of TM silica nanoparticles in 0.010 M NaCl.**

## Bibliography

1. Hamer, W.J. and Y.C. Wu, *Osmotic coefficients and mean activity coefficients of uni-univalent electrolytes in water at 25° C.* Journal of Physical and Chemical Reference Data, 1972. **1**(4): p. 1047-1100.

Supporting info article III:

Hofmeister effects in the gelling of silica nanoparticles in mixed salt solutions

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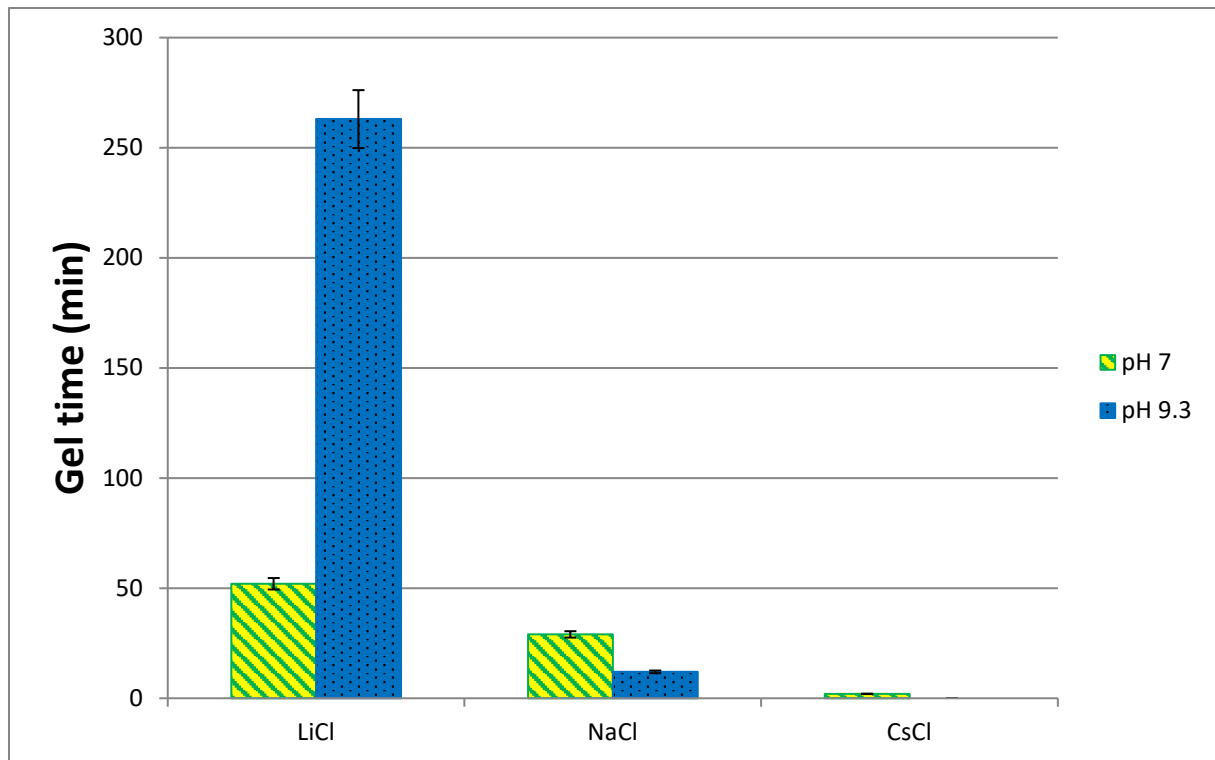
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Table S1: Debye lengths for the salt mixtures of MgCl<sub>2</sub> with monovalent chlorides as given in Figure 4.

Concentration monovalent ion (M)	Debye length (nm)
0	1.1148
0.0114	1.0740
0.0222	1.0200
0.0324	0.9749
0.0421	0.9363
0.0513	0.9050
0.0600	0.8775

Table S2: same as for Table S1 but for CaCl<sub>2</sub>.

Concentration monovalent ion (M)	Debye length (nm)
0	1.0200
0.0120	0.9706
0.0232	0.9294
0.0339	0.8955
0.0440	0.8661
0.0535	0.8412
0.0625	0.8197



Figur S1: Gel times for monovalent salts at 0.421M concentration at pH 7 and at pH 9.3. We do not observe pH dependent shift in gel time when going from strongly hydrated  $\text{Li}^+$  ion to weakly hydrated  $\text{Cs}^+$  ion as was obtained for divalent ions.

$[\text{Cl}^-]_{\text{TOT}} = 47.00 \text{ mM}$

$[\text{Mg}^{2+}]_{\text{TOT}} = 23.50 \text{ mM}$

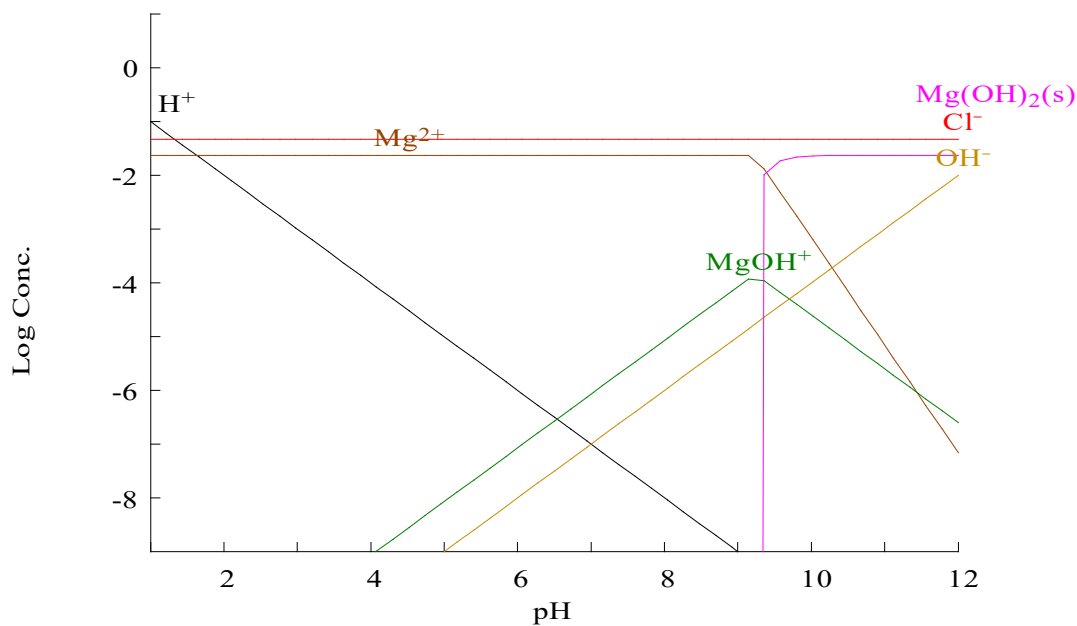


Figure S2: pH dependent logarithmic concentrations of the species present in  $\text{MgCl}_2$  solution. Note concentration used for these calculations are the same as used to obtain gel time results shown in Figure 5. The speciation diagram is generated by using Hydro Medusa.

$[\text{Cl}^-]_{\text{TOT}} = 47.00 \text{ mM}$

$[\text{Mg}^{2+}]_{\text{TOT}} = 23.50 \text{ mM}$

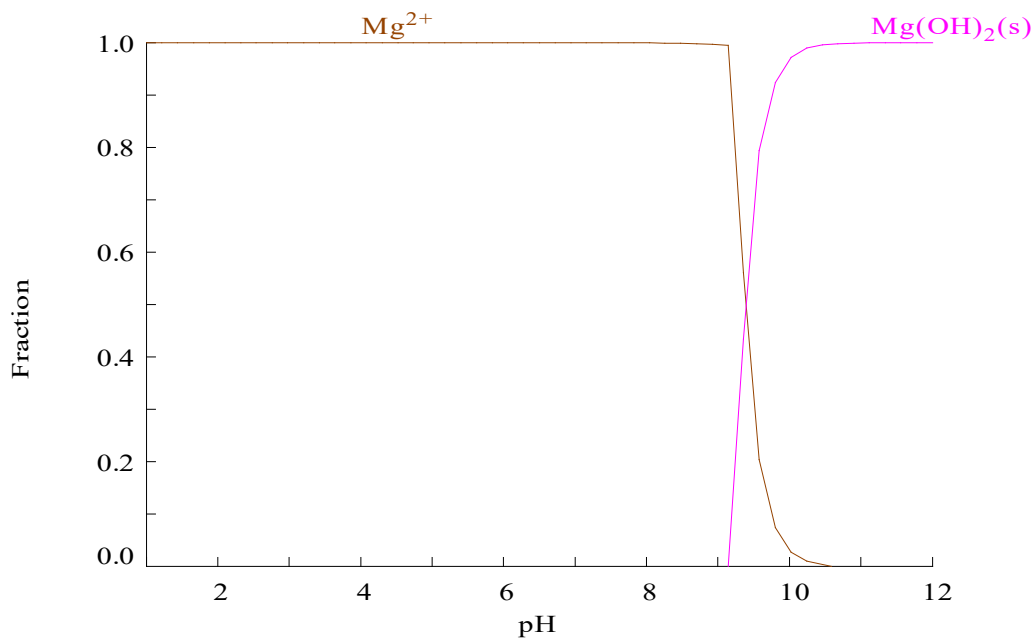


Figure S3: Same as in Figure S2 but for fractions of the species present in  $\text{MgCl}_2$  versus pH.



$[\text{Ca}^{2+}]_{\text{TOT}} = 29.60 \text{ mM}$

$[\text{Cl}^-]_{\text{TOT}} = 59.20 \text{ mM}$

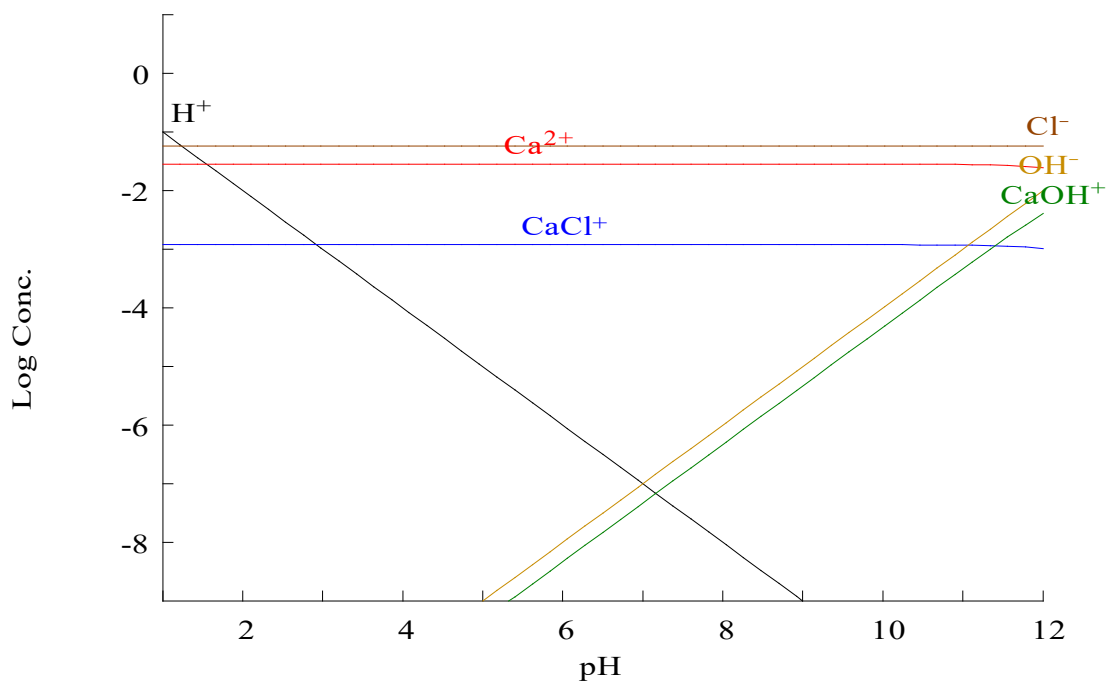


Figure S4: Same as in Figure S2 but for  $\text{CaCl}_2$ .

$[\text{Ca}^{2+}]_{\text{TOT}} = 29.60 \text{ mM}$

$[\text{Cl}^-]_{\text{TOT}} = 59.20 \text{ mM}$

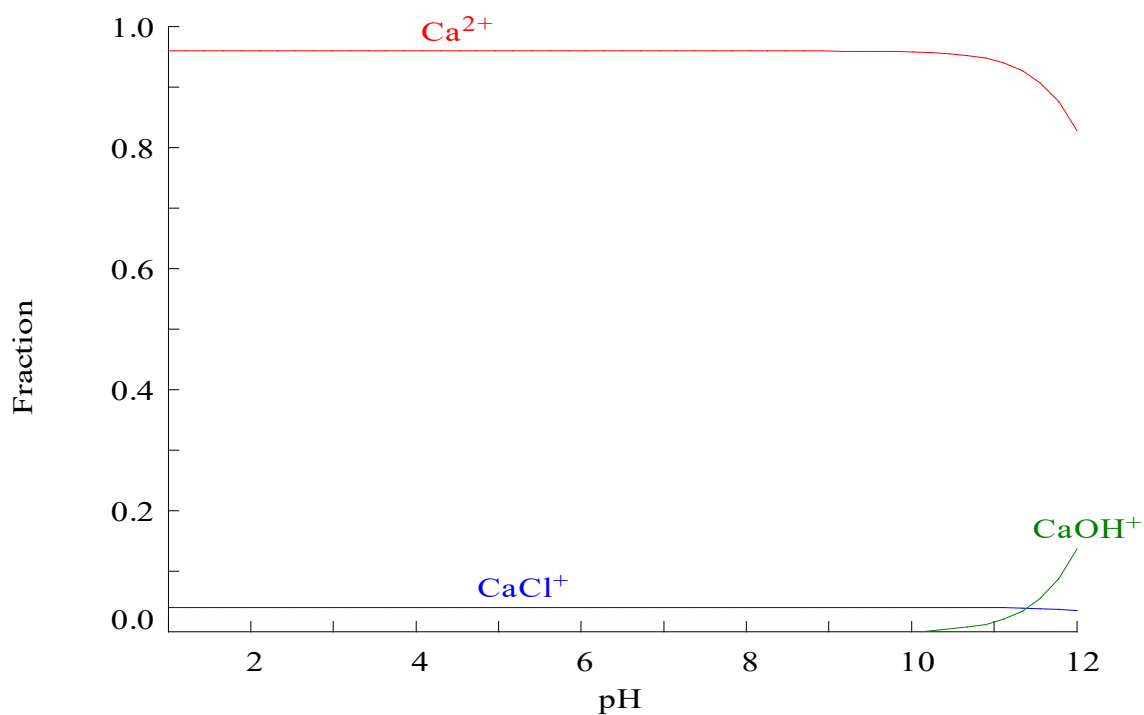


Figure S5: Same as in Figure S3 but for  $\text{CaCl}_2$ .

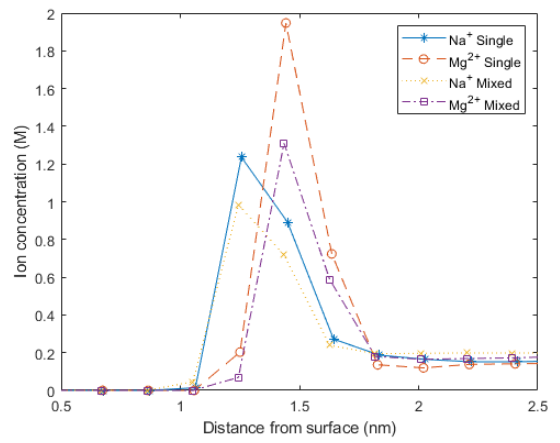
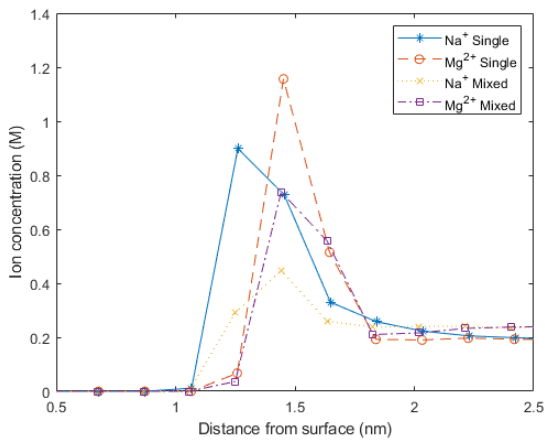


Figure S6: Ion distributions close to the silica surface for  $Mg^{2+}/Na^+$  mixtures and single salts. Left: pH 7. Right: pH 9.

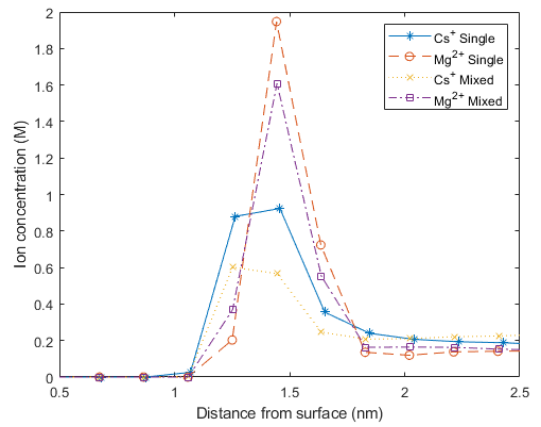
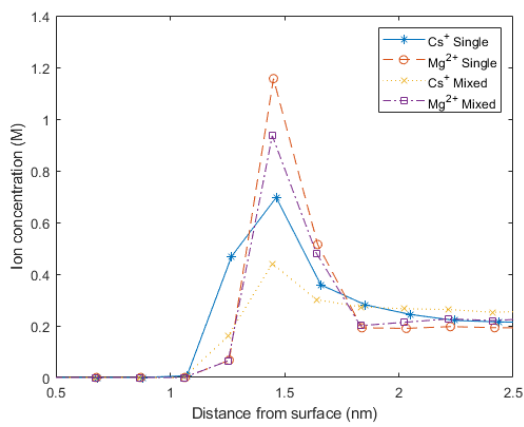


Figure S7: Ion distributions close to the silica surface for  $Mg^{2+}/Cs^+$  mixtures and single salts. Left: pH 7. Right: pH 9.

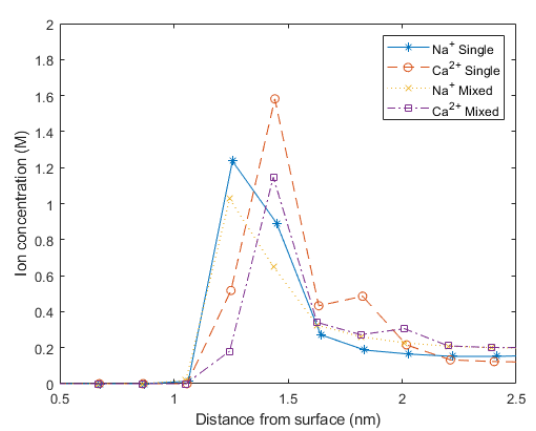
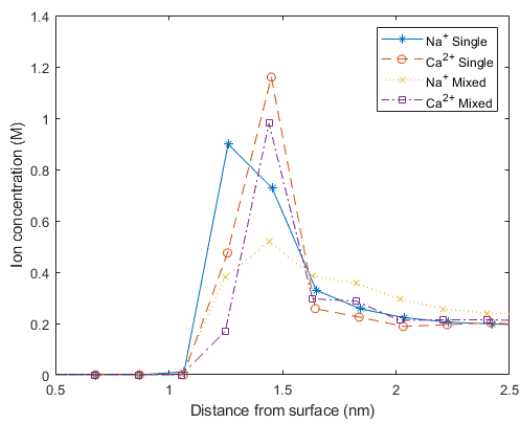


Figure S8: Ion distributions close to the silica surface for  $Ca^{2+}/Na^+$  mixtures and single salts. Left: pH 7. Right: pH 9.

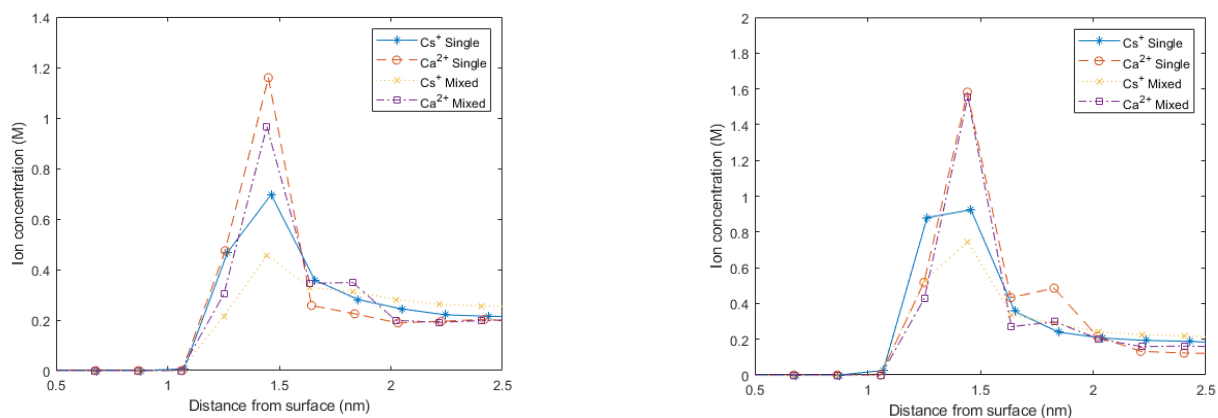


Figure S9: Ion distributions close to the silica surface for  $\text{Ca}^{2+}/\text{Cs}^+$  mixtures and single ions. Left: pH 7. Right: pH 9.

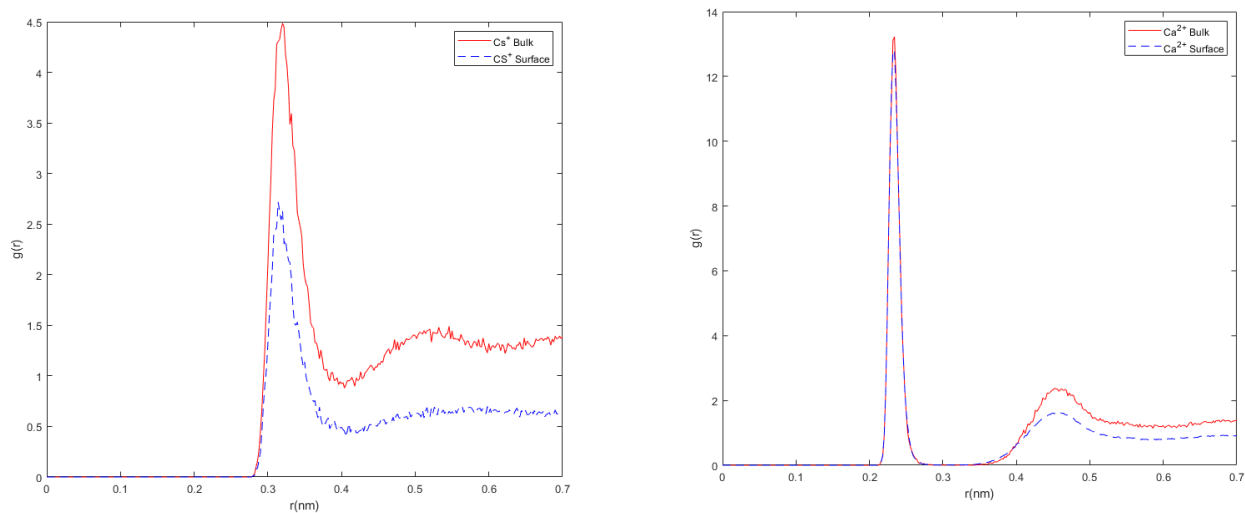


Figure S10: Left: Radial distribution of water molecules surrounding the  $\text{Cs}^+$  ions in the simulation cell.  $\text{Cs}^+$  Bulk refers to the distribution of water around ions far  $\approx 4$  nm from the surface.  $\text{Cs}^+$  Surface refers to the distribution around  $\text{Cs}^+$  ions situated at the silica surface ( $0.2 \text{ nm} \pm 0.2 \text{ nm}$  from the surface). Right: Radial distribution of water molecules surrounding the  $\text{Ca}^{2+}$  ions in the simulation cell.  $\text{Ca}^{2+}$  Bulk refers to the distribution of water around ions far  $\approx 4$  nm from the surface.  $\text{Ca}^{2+}$  Surface refers to the distribution around  $\text{Ca}^{2+}$  ions situated at the silica surface ( $0.6 \text{ nm} \pm 0.2 \text{ nm}$  from the surface).

Supporting info article IV:

## Supporting Information

# Development and Evaluation of Polyether Ether Ketone (PEEK) Capillary for Electrospray

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\* Corresponding Author

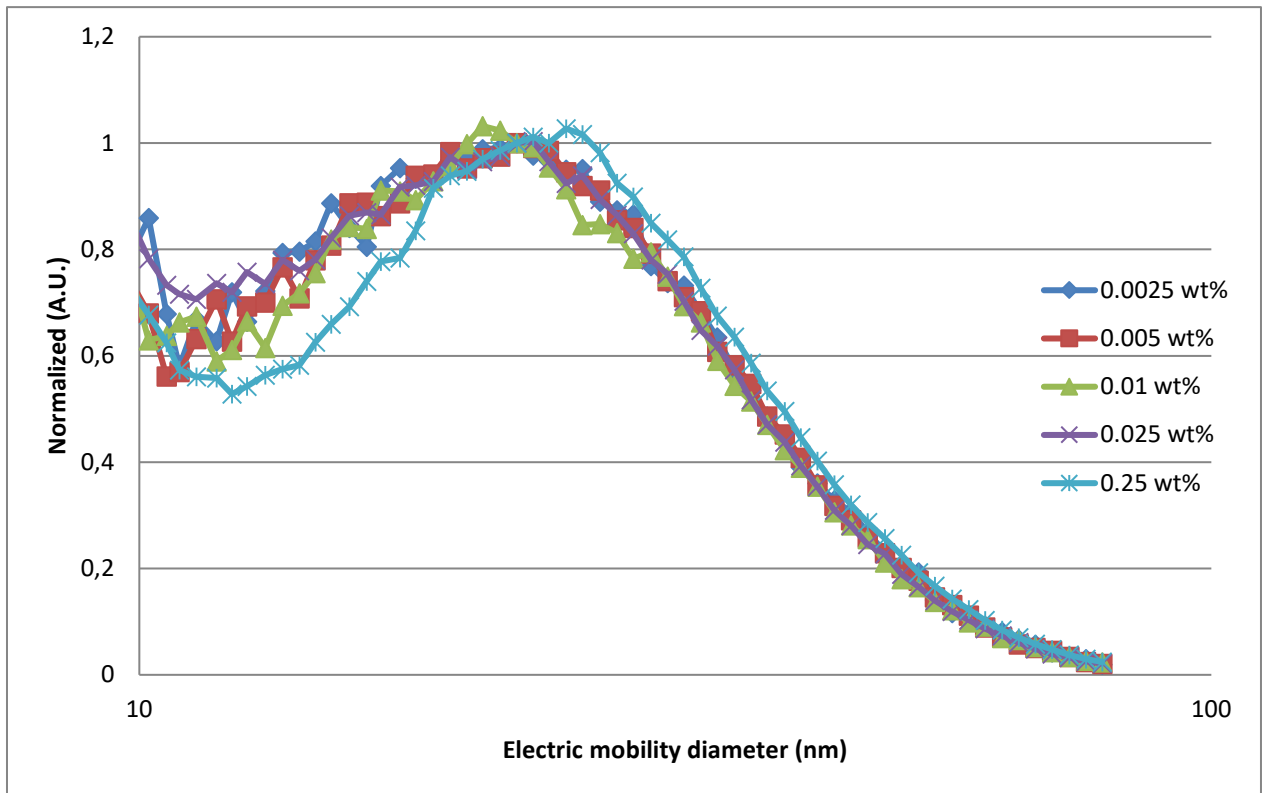


Figure S8: Size distributions for a number of silica particle concentrations. All concentrations <0.25 wt% show no clear shift while the 0.25 wt% distribution show a minor shift towards larger size distribution. This suggests that the concentration at 0.25 wt% is high enough to start generating more than one particle per sprayed droplet which is not desirable.

Supporting info article V:

## 1. Activation Energies

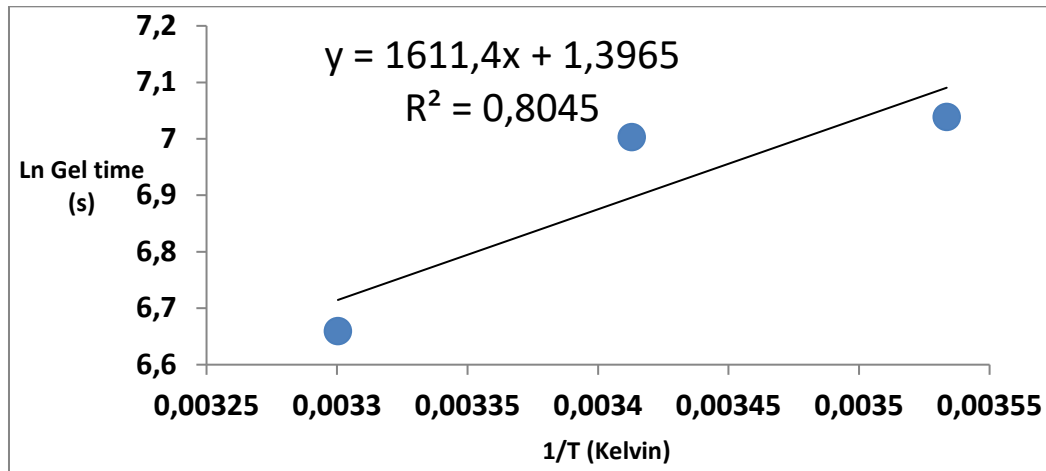


Figure S9: Graph for calculation of activation energy in this case for aggregation of CS40-236. The slope of the inserted trend line is equal to  $E_a/R$ .

## 2. ES-SMPS Results

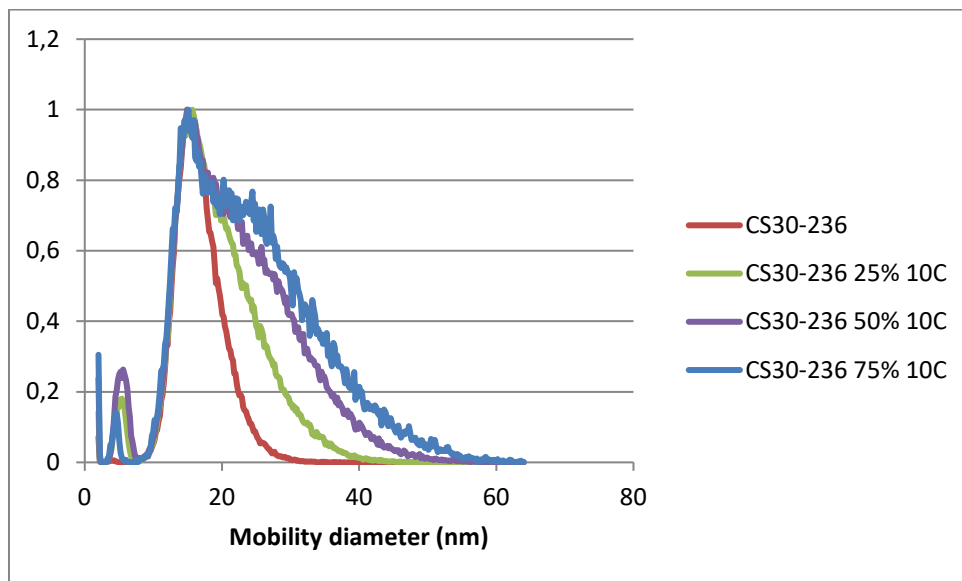


Figure S10: Normalized particle size distribution for CS30-236 gelling at 10°C.

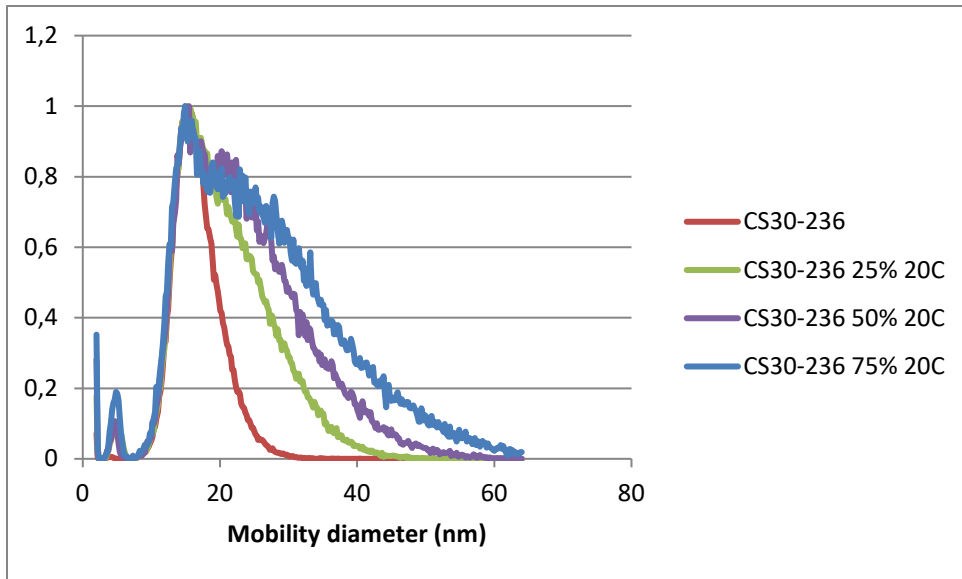


Figure S11: Normalized particle size distribution for CS30-236 gelling at 20°C.

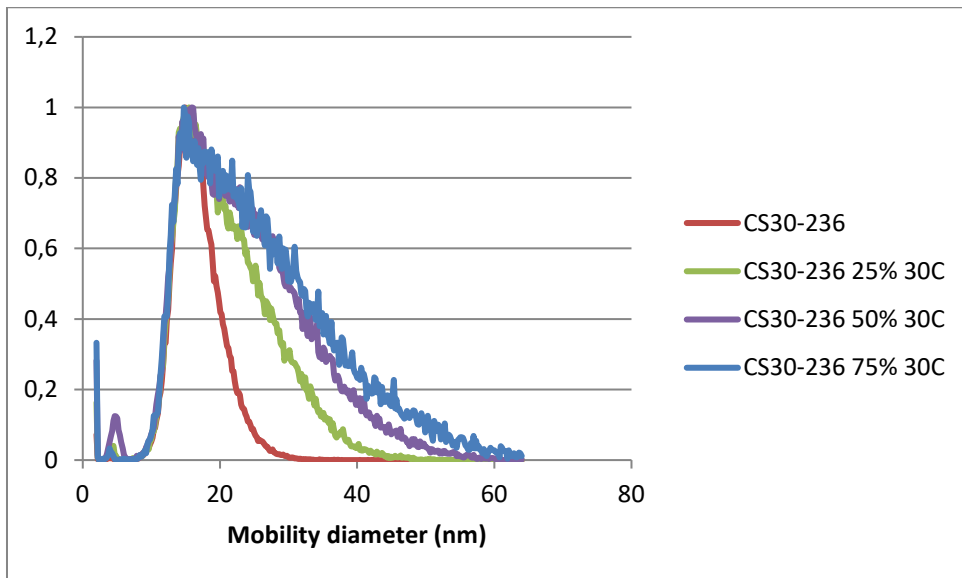


Figure S12: Normalized particle size distribution for CS30-236 gelling at 30°C.

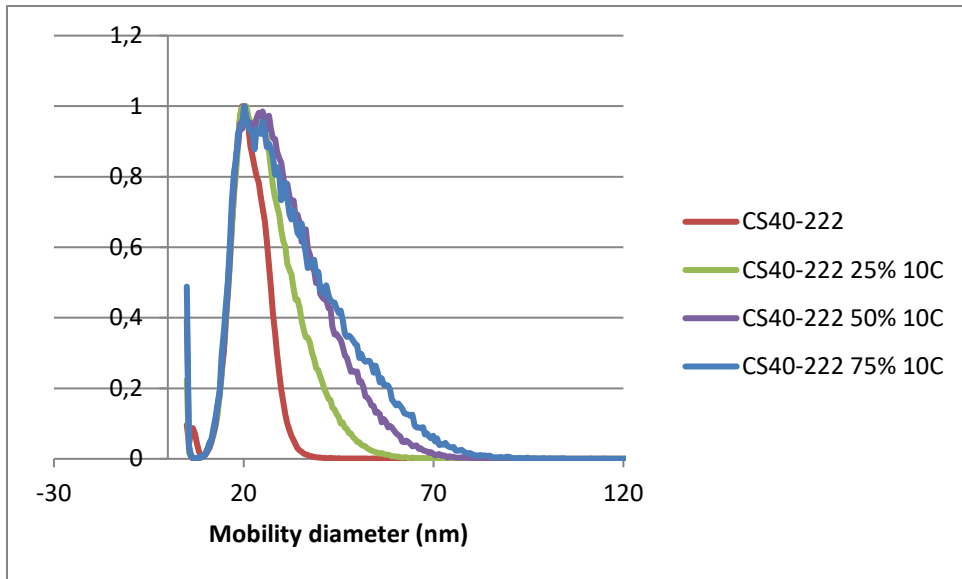


Figure S13: Normalized particle size distribution for CS40-222 gelling at 10°C.

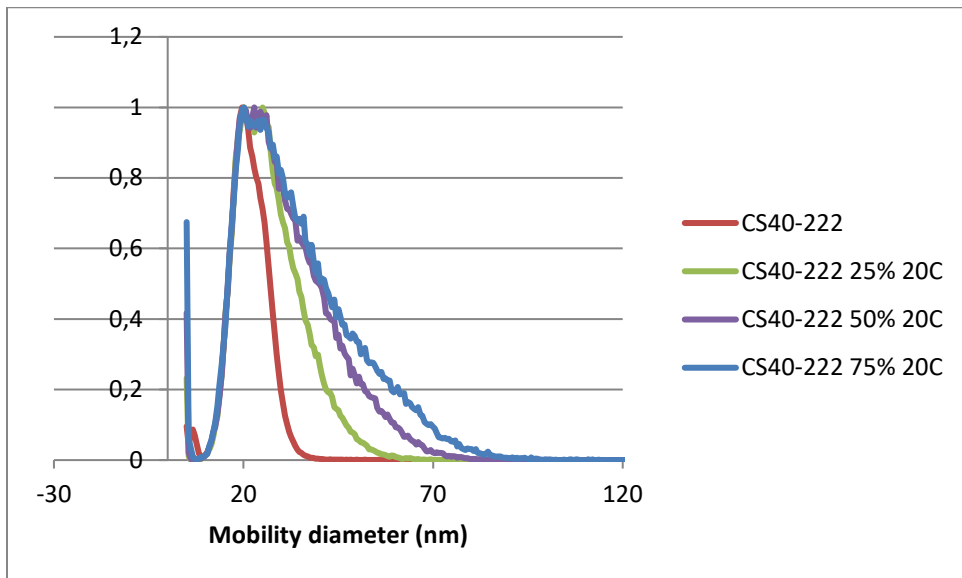


Figure S14: Normalized particle size distribution for CS40-222 gelling at 20°C.



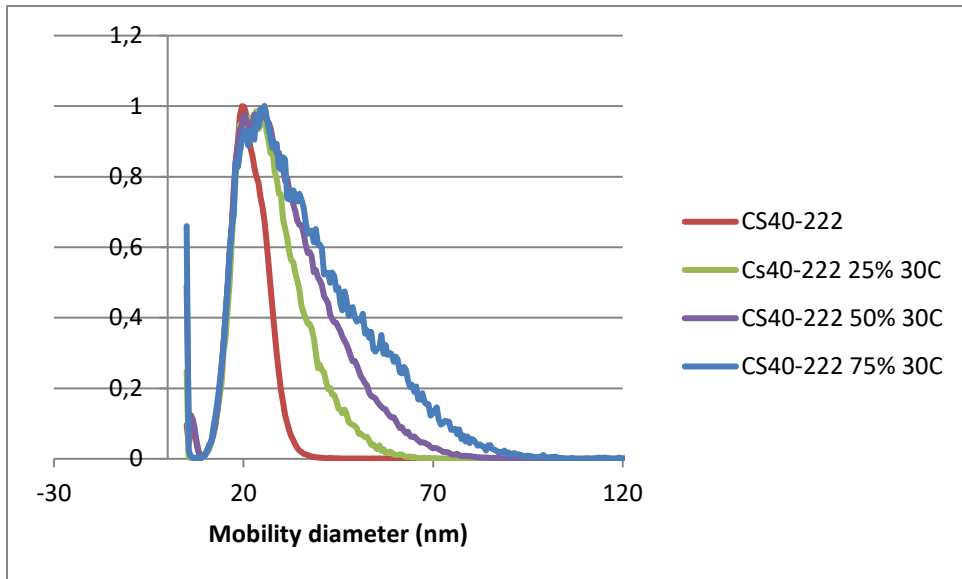


Figure S15: Normalized particle size distribution for CS40-222 gelling at 30°C.

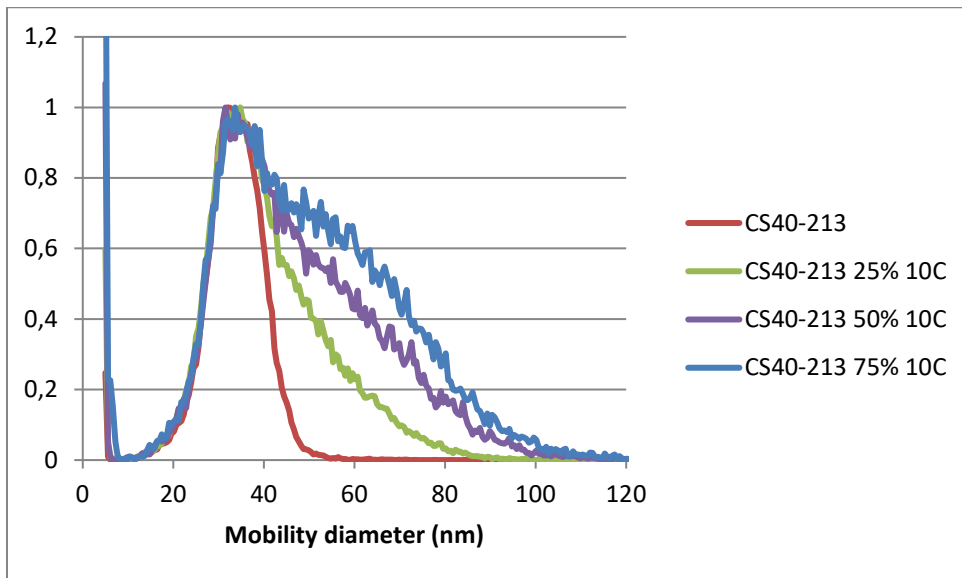


Figure S16: Normalized particle size distribution for CS40-213 gelling at 10°C.

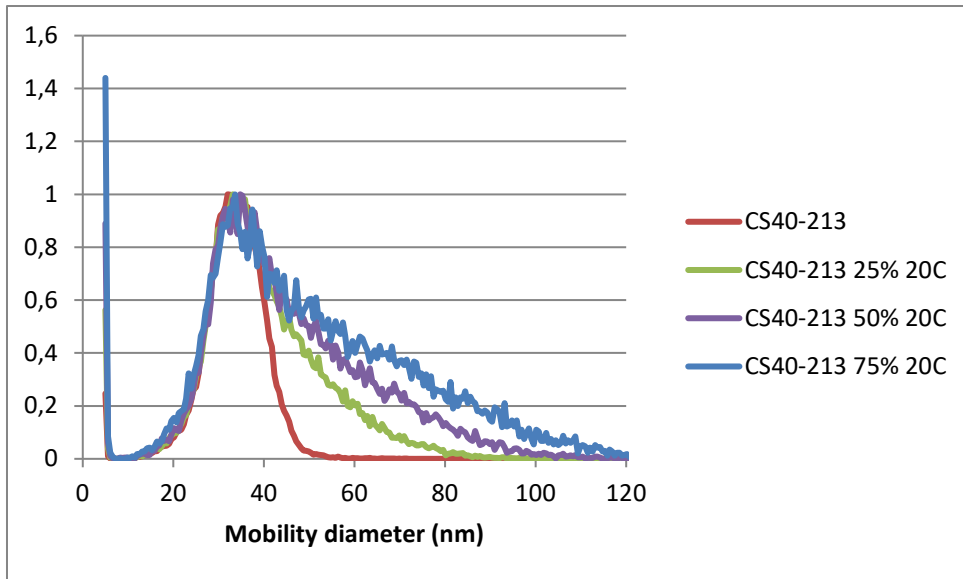


Figure S17: Normalized particle size distribution for CS40-213 gelling at 20°C.

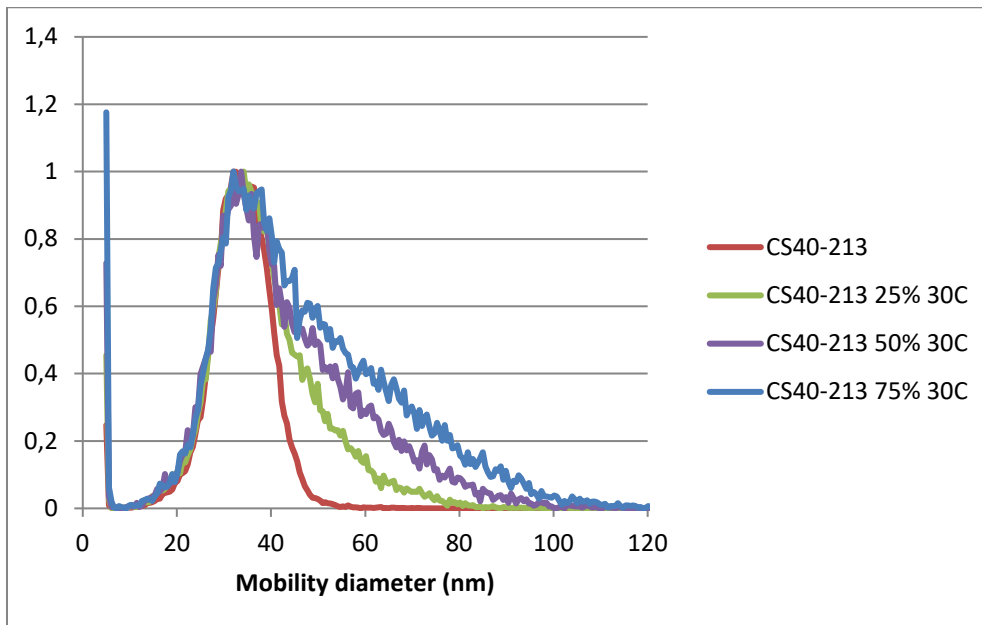


Figure S18: Normalized particle size distribution for CS40-213 gelling at 30°C.

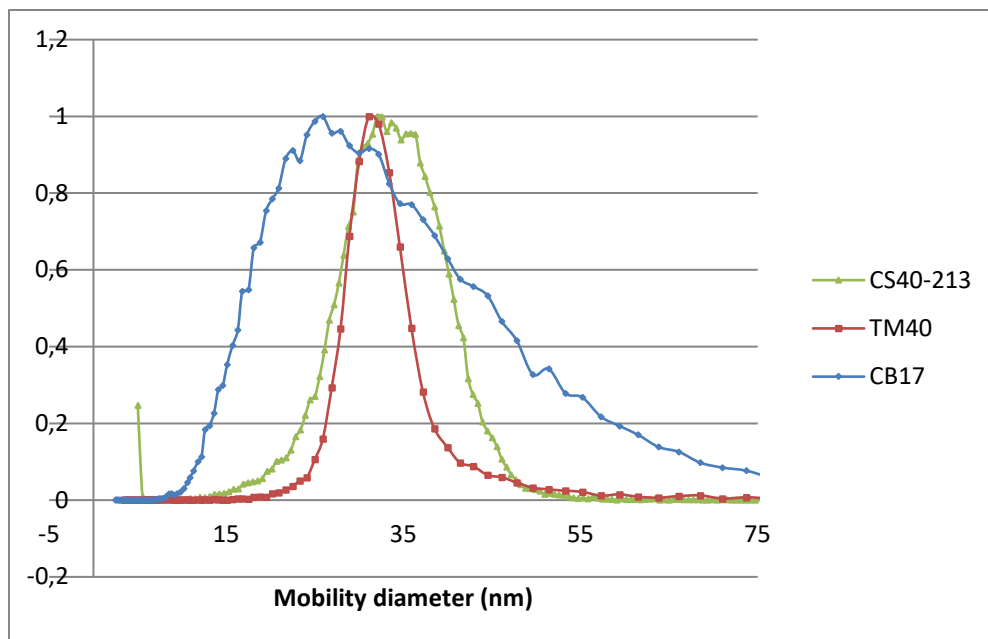


Figure S19: Normalized particle size distribution for TM40, CB17, and CS40-213 silica sols. These distributions are valid for the sols before aggregation is initiated.

Table S1: The results of the ES-SMPS measurements for CS30-236 sol. The  $N_d$  is also included as calculated from equation 1 using the number average particle sizes. The % gelled is with reference to the gel time for each temperature from Error! Reference source not found. by which the sample has been taken from the aggregating sol and fed into the ES-SMPS, with the corresponding time shown as well under Time.

Sol	Temp °C	% gelled	Time (min)	Number average particle size (nm)	$N_d$ (calculated from equation 1)
CS30-236	Ambient	0	0	16.82	1.00
CS30-236	10	25	4.75	19.78	1.41
CS30-236	10	50	9.50	22.64	1.87
CS30-236	10	75	14.25	24.64	2.23
CS30-236	20	25	4.58	21.03	1.60
CS30-236	20	50	9.17	23.75	2.06
CS30-236	20	75	13.75	26.61	2.62
CS30-236	30	25	3.25	21.16	1.62
CS30-236	30	50	6.50	24.02	2.11
CS30-236	30	75	9.75	26.08	2.51

Table S2: The results of the ES-SMPS measurements for CS40-222 sol. The  $N_d$  is also included as calculated from equation 1 using the number average particle sizes. The % gelled is with reference to the gel time for each temperature from Error!

Reference source not found. by which the sample has been taken from the aggregating sol and fed into the ES-SMPS, with the corresponding time shown as well under Time.

Sol	Temp °C	% gelled	Time (min)	Number average particle size (nm)	N <sub>d</sub> (calculated from equation 1)
CS40-222	Ambient	0	0	21.67	1.00
CS40-222	10	25	25.25	26.81	1.56
CS40-222	10	50	50.50	31.05	2.13
CS40-222	10	75	75.75	33.56	2.51
CS40-222	20	25	19.92	27.25	1.62
CS40-222	20	50	39.84	31.58	2.21
CS40-222	20	75	59.75	34.72	2.69
CS40-222	30	25	13.08	27.74	1.68
CS40-222	30	50	26.17	31.94	2.26
CS40-222	30	75	39.25	36.84	3.04

**Table S3: The results of the ES-SMPS measurements for CS40-213 sol. The N<sub>d</sub> is also included as calculated from equation 1 using the number average particle sizes. The % gelled is with reference to the gel time for each temperature from Error! Reference source not found. by which the sample has been taken from the aggregating sol and fed into the ES-SMPS, with the corresponding time shown as well under Time.**

Sol	Temp °C	% gelled	Time (min)	Number average particle size (nm)	N <sub>d</sub> (calculated from equation 1)
CS40-213	Ambient	0	0	33.78	1.00
CS40-213	10	25	166.33	41.41	1.53
CS40-213	10	50	332.67	48.03	2.09
CS40-213	10	75	479.00	51.08	2.38
CS40-213	20	25	105.17	40.83	1.49
CS40-213	20	50	210.34	46.48	1.95
CS40-213	20	75	315.50	52.56	2.53
CS40-213	30	25	51.83	39.00	1.35
CS40-213	30	50	103.67	43.94	1.74
CS40-213	30	75	155.50	48.25	2.11

Supporting info article VI:

## The long term stability of silica nanoparticle gels in waters of different ionic compositions and pH values

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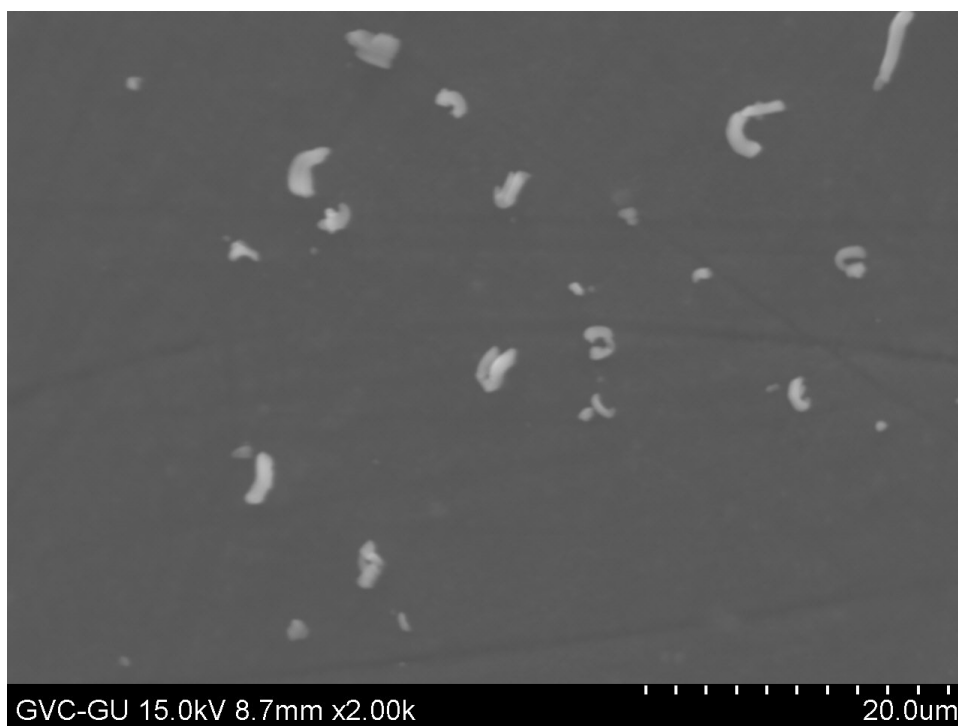
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### Supporting material

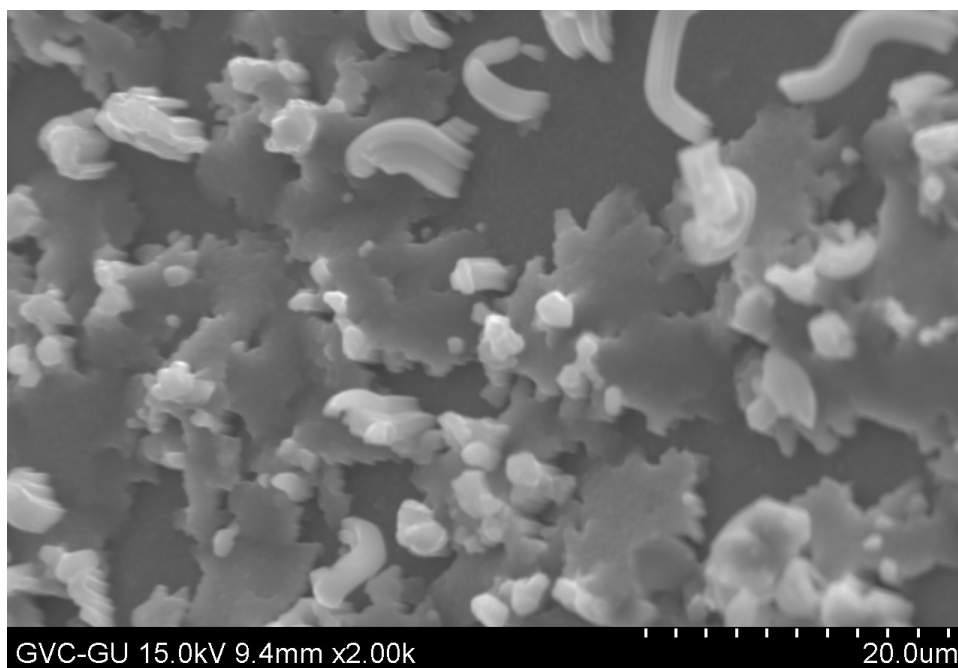
#### Scanning Electron Microscope Pictures:

To produce the Scanning electron microscope (SEM) pictures small pieces of gel (approximately 1x1x0.1 cm) were dried in a vacuum oven at 70°C overnight. The pieces were then sputtered with a thin layer of carbon to increase the conductivity of the surface.

SEM images are shown below in Figure S1-S3. Figure S1 is for a newly formed gel which has been dried within 24 h of gelling. As an accelerator, KCl was used and the gel is made from the same sol and had the same gel time as was in the long term stability tests. It is apparent from the figure that the gel surface is smooth and as can be expected for a new gel, it shows no signs of corrosion or dissolution. The bright spots are of KCl crystals (confirmed by SEM-EDX) formed during drying of the gel. These crystals have probably formed over pore openings in the gel and are more clearly seen in Figure S2.



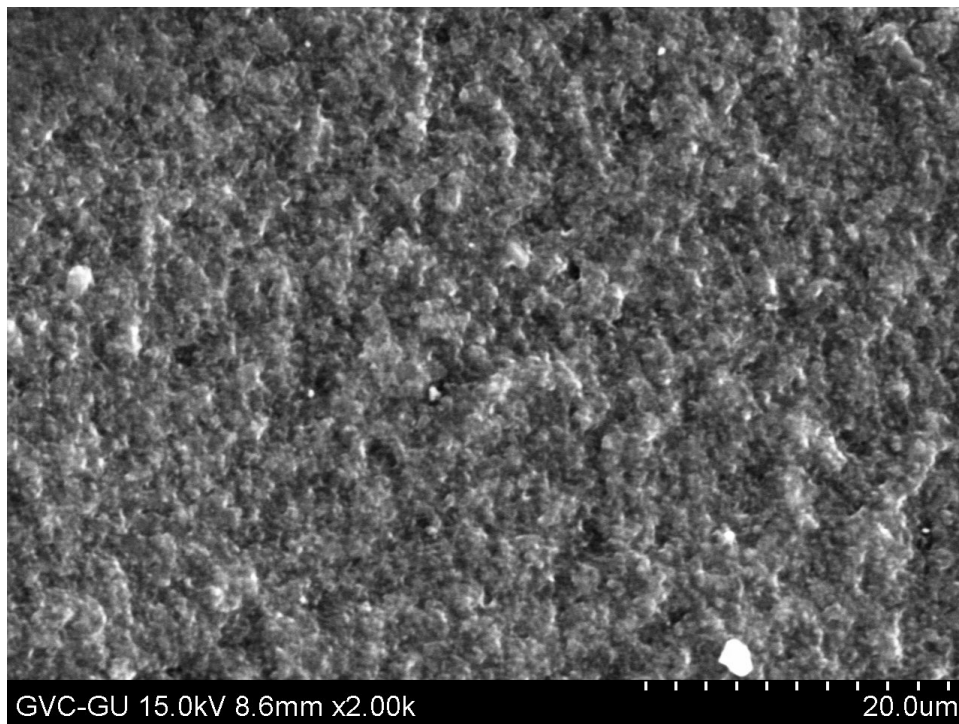
*Figure S7: Surface of silica gel formed with KCl accelerator and dried within 24 h of forming. The surface is smooth with a few potassium chloride crystals on the surface.*



*Figure S8: Here the potassium chloride crystals are clearly visible on the newly formed silica surface. The crystals have formed during drying as water has exited small pores in the silica surface.*

In Figure S3 the surface of a gel from setup F is shown. Setup F had a mixture of ions (108 mM Na<sup>+</sup>, 34 mM Ca<sup>2+</sup>, 5 mM Mg<sup>2+</sup>) in the test water (pH 12) passing through the

gels and the setup was run for a total of 221 days. The surface of the gel is coarser than the newly formed gel which is a clear sign of corrosion. It is not unexpected for a gel surface that has been exposed to solution having pH 12 for an extended time that the surface shows signs of dissolution. As we discussed in the paper the dissolved silica might well have re-precipitated inside the gel body as the pH changed to 10. Figure S3 might thus show the top of the transition layer.



*Figure S9: Surface of silica that has been exposed to water in the stability tests. This is silica from the top layer of setup F. As can be seen the silica surface is coarse and seems to have been corroded by the water. Also there was a lack of salt crystals on the surface compared to the new gel in Figure S1 and S2.*