## SUPPORTING INFORMATION

# Synthesis, polymerization and assembly of nano-silica particles below the isoelectric point 

Alberto Lazaro ${ }^{1 *}$, Neus Vilanova ${ }^{2}$, Luana. D. Barreto Torres ${ }^{1}$, Gea Resoort ${ }^{1}$, Ilja. K. Voets ${ }^{2}$, H.J.H. Brouwers ${ }^{1}$

1) Department of the Built Environment, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands
2) Department of Chemical Engineering and Chemistry, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands
(*) To whom correspondence should be addressed: tel: +31 (0)40 247 2006, e-mail: a.lazaro.garcia@tue.nl

## Contents:

Figure S1 Particle size distribution of the grounded olivine.
Figure S2. TEM pictures of nano-silica produced below the IEP.
Figure S3. Normal size distribution of silica particles measured in Figure S2.
Figure S4. TEM pictures of nano-silica produced below the IEP.
Figure S5. Normal size distribution of silica particles measured in Figure S4.
Figure S6. TEM pictures of nano-silica produced below the IEP.
Figure S7. Normal size distribution of silica particles measured in Figure S6.

Table S1. Estimation of the number of particles present in the system at the CAC.
Table S2. Estimated particle size of silica particles.
Table S3. Estimated specific surface area of fully condensed particles of the size determined by laser light scattering.
Table S4. Specific surface area and BJH pore volume for silicas produced below the IEP at different reaction conversions (X).


Figure S1. Particle size distribution of the grounded mineral olivine analyzed with a Malvern 2000 device. The particle size is in the range 0.8 to $500 \mu \mathrm{~m}$.


Figure S2. TEM pictures of nano-silica produced below the IEP (Experiment NS-2 [23]). More than 30 primary particles diameters were measured using the ImageJ software.


Figure S3. Normal size distribution of primary silica particles measured in Figure S2.


Figure S4. TEM pictures of nano-silica produced below the IEP (Experiment NS-2 [23]). More than 30 primary particles diameters were measured using the ImageJ software.


Figure S5. Normal size distribution of primary silica particles measured in Figure S4.


Figure S6. TEM pictures of nano-silica produced below the IEP (Experiment NS-2 [23]). More than 30 silica aggregates were measured using the ImageJ software.


Figure S7. Normal size distribution of silica aggregates measured in Figure S6.

Table S1. Estimation of the number of particles present in the system at the CAC. Where $[\mathrm{Si}]_{\mathrm{CAC}}$ is the critical aggregation concentration of silica, $\mathrm{D}_{1 \mathrm{P}}$ the particle diameter at the $C A C, V_{1 P}$ the volume of one particle, $m_{1 P}$ the mass of one particle and $n_{P}$ the number of particles at the CAC.

| $\rho_{\mathrm{Si}}$ | 2.21 | $\mathrm{~kg} / \mathrm{L}$ |
| :---: | :---: | :---: |
| $\mathrm{MW}_{\mathrm{SiO} 2}$ | 60.00 | $\mathrm{~g} / \mathrm{mol}$ |
| $[\mathrm{Si}]_{\mathrm{CAC}}$ | 30 | mM |
| $\mathrm{m}_{\mathrm{Si}}$ | 1.80 | $\mathrm{~g} / \mathrm{L}$ |
| $\mathrm{D}_{1 \mathrm{P}}$ | 3.0 | nm |
| $\mathrm{~V}_{1 \mathrm{P}}$ | $1.41 \mathrm{E}-23$ | L |
| $\mathrm{~m}_{1 \mathrm{P}}$ | $3.12 \mathrm{E}-20$ | g |
| $\mathrm{n}_{\mathrm{P}}$ | $5.76 \mathrm{E}+19$ | particles $/ \mathrm{L}$ |

Table S2. Estimated particle size of silica particles. It is assumed in this calculation that the number of particles remains constant and the silica particles grow through the condensation mechanism forming fully condensed spheres. $V_{T}$ is the total volume of silica per liter of solution.

| $[\mathbf{S i}]$ <br> $(\mathbf{m M})$ | $\mathbf{m}_{\mathbf{S i}}$ <br> $(\mathbf{g} / \mathbf{L})$ | $\mathbf{V}_{\mathbf{T}}$ <br> $(\mathbf{L} / \mathbf{L})$ | $\mathbf{V}_{\mathbf{1 P}}$ <br> $(\mathbf{L})$ | $\mathbf{D}_{\mathbf{1}}$ <br> $(\mathbf{n m})$ |
| :---: | :---: | :---: | :---: | :---: |
| 30 | 1.80 | $8.14 \mathrm{E}-04$ | $1.41 \mathrm{E}-23$ | 3.00 |
| 35 | 2.10 | $9.50 \mathrm{E}-04$ | $1.65 \mathrm{E}-23$ | 3.16 |
| 40 | 2.40 | $1.09 \mathrm{E}-03$ | $1.88 \mathrm{E}-23$ | 3.30 |
| 45 | 2.70 | $1.22 \mathrm{E}-03$ | $2.12 \mathrm{E}-23$ | 3.43 |
| 50 | 3.00 | $1.36 \mathrm{E}-03$ | $2.36 \mathrm{E}-23$ | 3.56 |
| 55 | 3.30 | $1.49 \mathrm{E}-03$ | $2.59 \mathrm{E}-23$ | 3.67 |
| 60 | 3.60 | $1.63 \mathrm{E}-03$ | $2.83 \mathrm{E}-23$ | 3.78 |
| 65 | 3.90 | $1.76 \mathrm{E}-03$ | $3.06 \mathrm{E}-23$ | 3.88 |
| 70 | 4.20 | $1.90 \mathrm{E}-03$ | $3.30 \mathrm{E}-23$ | 3.98 |
| 75 | 4.50 | $2.04 \mathrm{E}-03$ | $3.53 \mathrm{E}-23$ | 4.07 |
| 80 | 4.80 | $2.17 \mathrm{E}-03$ | $3.77 \mathrm{E}-23$ | 4.16 |
| 85 | 5.10 | $2.31 \mathrm{E}-03$ | $4.01 \mathrm{E}-23$ | 4.25 |
| 90 | 5.40 | $2.44 \mathrm{E}-03$ | $4.24 \mathrm{E}-23$ | 4.33 |
| 95 | 5.70 | $2.58 \mathrm{E}-03$ | $4.48 \mathrm{E}-23$ | 4.41 |
| 100 | 6.00 | $2.71 \mathrm{E}-03$ | $4.71 \mathrm{E}-23$ | 4.48 |
| 105 | 6.30 | $2.85 \mathrm{E}-03$ | $4.95 \mathrm{E}-23$ | 4.55 |
| 110 | 6.60 | $2.99 \mathrm{E}-03$ | $5.18 \mathrm{E}-23$ | 4.63 |
| 115 | 6.90 | $3.12 \mathrm{E}-03$ | $5.42 \mathrm{E}-23$ | 4.70 |

Table S3. Estimated specific surface area of fully condensed particles of the size determined by laser light scattering. It is assumed in this calculation that fully condensed particles spherical particles are formed of the sizes determined by DLS.

| $[\mathbf{S i}]$ <br> $(\mathbf{m M})$ | $\mathbf{D}_{\mathbf{1 P}}$ <br> $(\mathbf{n m})$ | $\mathbf{m}_{\mathbf{S i}}$ <br> $(\mathbf{g} / \mathbf{L})$ | $\mathbf{V}_{\mathbf{T}}$ <br> $(\mathbf{L} / \mathbf{L})$ | $\mathbf{V}_{\mathbf{1 P}}$ <br> $(\mathbf{L})$ | $\mathbf{n}_{\mathbf{P}}$ | $\mathbf{S S A}$ <br> $\left(\mathbf{m}^{2} / \mathbf{g}\right)$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| 30 | 3.0 | 1.80 | $8.14 \mathrm{E}-04$ | $1.41 \mathrm{E}-23$ | $5.76 \mathrm{E}+19$ | 905.0 |
| 35 | 7.3 | 2.10 | $9.50 \mathrm{E}-04$ | $2.01 \mathrm{E}-22$ | $4.74 \mathrm{E}+18$ | 373.8 |
| 40 | 14.3 | 2.40 | $1.09 \mathrm{E}-03$ | $1.54 \mathrm{E}-21$ | $7.03 \mathrm{E}+17$ | 189.3 |
| 45 | 22.9 | 2.70 | $1.22 \mathrm{E}-03$ | $6.31 \mathrm{E}-21$ | $1.94 \mathrm{E}+17$ | 118.4 |
| 50 | 33.0 | 3.00 | $1.36 \mathrm{E}-03$ | $1.89 \mathrm{E}-20$ | $7.20 \mathrm{E}+16$ | 82.2 |
| 55 | 44.6 | 3.30 | $1.49 \mathrm{E}-03$ | $4.66 \mathrm{E}-20$ | $3.21 \mathrm{E}+16$ | 60.8 |
| 60 | 57.8 | 3.60 | $1.63 \mathrm{E}-03$ | $1.01 \mathrm{E}-19$ | $1.62 \mathrm{E}+16$ | 47.0 |
| 65 | 72.4 | 3.90 | $1.76 \mathrm{E}-03$ | $1.99 \mathrm{E}-19$ | $8.89 \mathrm{E}+15$ | 37.5 |
| 70 | 88.5 | 4.20 | $1.90 \mathrm{E}-03$ | $3.63 \mathrm{E}-19$ | $5.23 \mathrm{E}+15$ | 30.7 |
| 75 | 106.2 | 4.50 | $2.04 \mathrm{E}-03$ | $6.26 \mathrm{E}-19$ | $3.25 \mathrm{E}+15$ | 25.6 |
| 80 | 125.3 | 4.80 | $2.17 \mathrm{E}-03$ | $1.03 \mathrm{E}-18$ | $2.11 \mathrm{E}+15$ | 21.7 |
| 85 | 146.0 | 5.10 | $2.31 \mathrm{E}-03$ | $1.63 \mathrm{E}-18$ | $1.42 \mathrm{E}+15$ | 18.6 |
| 90 | 168.2 | 5.40 | $2.44 \mathrm{E}-03$ | $2.49 \mathrm{E}-18$ | $9.81 \mathrm{E}+14$ | 16.1 |
| 95 | 191.9 | 5.70 | $2.58 \mathrm{E}-03$ | $3.70 \mathrm{E}-18$ | $6.98 \mathrm{E}+14$ | 14.2 |
| 100 | 217.1 | 6.00 | $2.71 \mathrm{E}-03$ | $5.35 \mathrm{E}-18$ | $5.07 \mathrm{E}+14$ | 12.5 |

Table S4. Specific surface area and BJH pore volume for silicas produced below the IEP at different reaction conversions ( $\mathbf{X}$ ).

| $\mathbf{X}$ <br> $(\mathbf{\%})$ | $\mathbf{S S A}_{\mathbf{B E T}}$ <br> $\left(\mathbf{m}^{\mathbf{2}} / \mathbf{g}\right)$ | $\mathbf{V}_{\mathbf{P}^{\prime} \mathbf{A}}$ <br> $\left.\mathbf{B J H}^{\mathbf{3}} / \mathbf{g}\right)$ |
| :---: | :---: | :---: |
| 38 | 517 | 0.992 |
| 77 | 325 | 0.597 |
| 90 | 310 | 0.475 |

