

Calcination-free production of calcium hydroxide at sub-boiling temperatures

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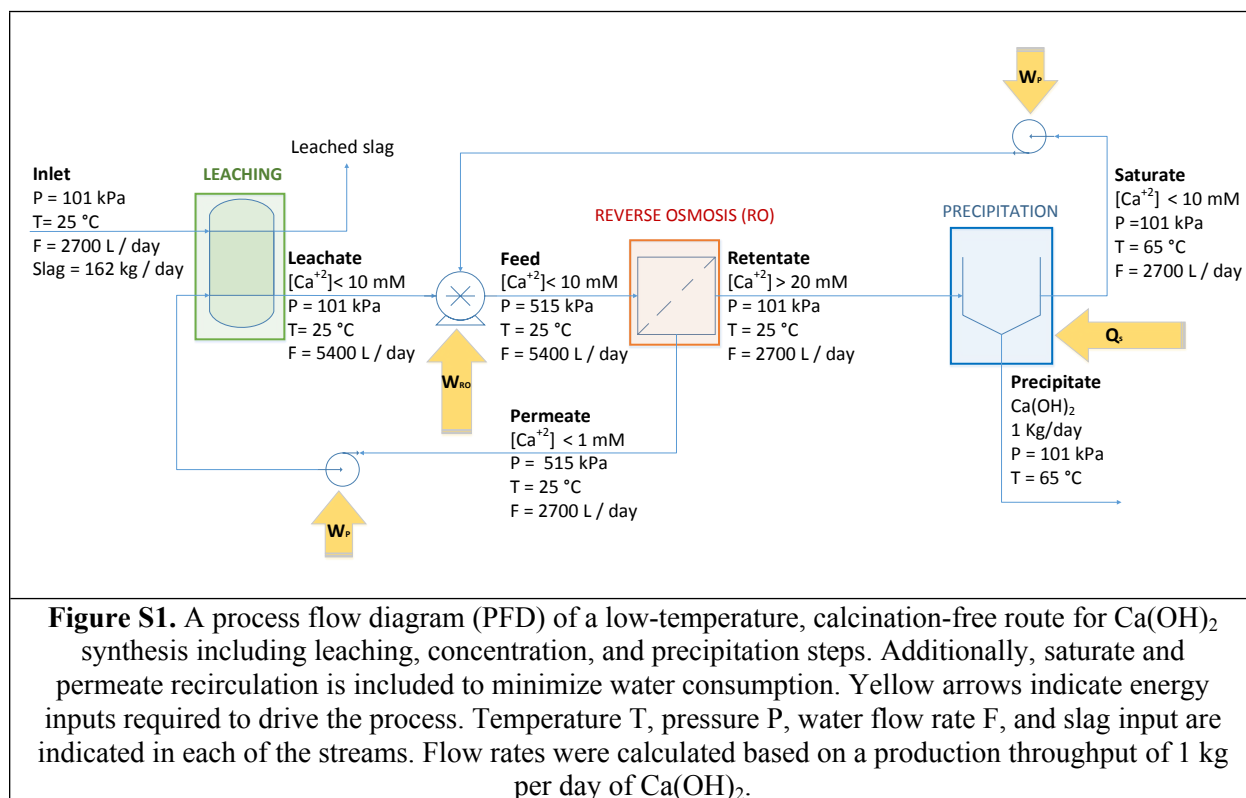


Figure S1. A process flow diagram (PFD) of a low-temperature, calcination-free route for Ca(OH)₂ synthesis including leaching, concentration, and precipitation steps. Additionally, saturate and permeate recirculation is included to minimize water consumption. Yellow arrows indicate energy inputs required to drive the process. Temperature T, pressure P, water flow rate F, and slag input are indicated in each of the streams. Flow rates were calculated based on a production throughput of 1 kg per day of Ca(OH)₂.

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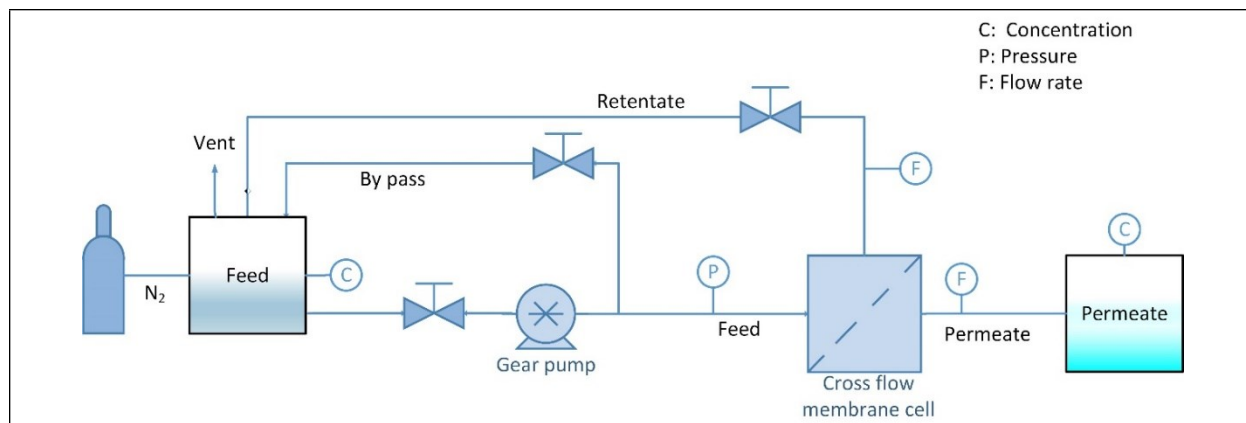


Figure S2. A schematic of the RO concentration setup showing cross flow membrane cell, feed, and permeate tanks, gear pump and streams configuration (retentate, permeate, bypass, and nitrogen lines). The circled C, P, and F indicate the location of concentration, pressure, and flow rate measurement sensors, respectively.

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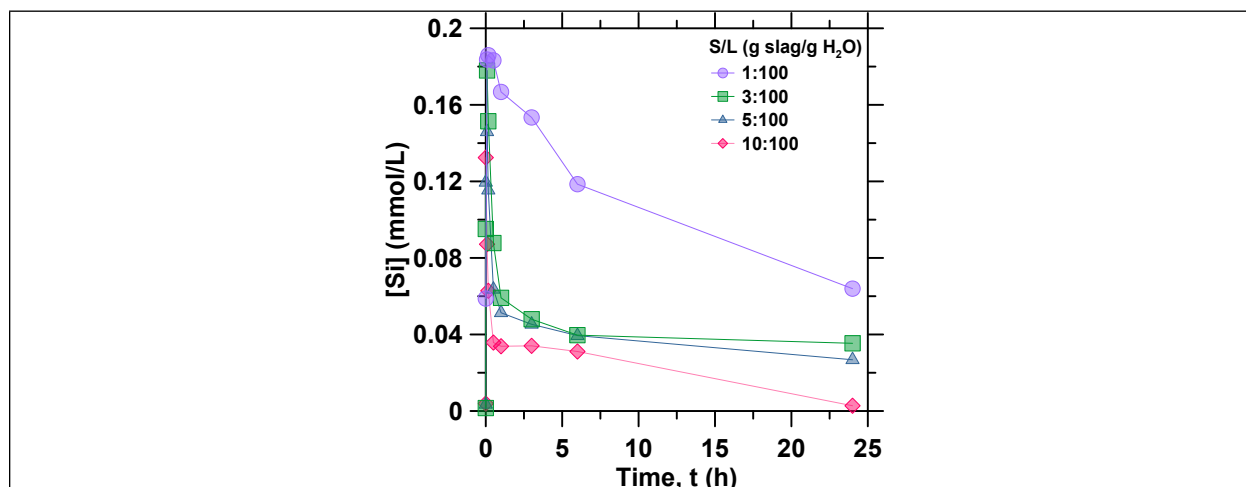


Figure S3. The silicon concentration as a function of time in stirred conditions following leaching of a BOF-slag.

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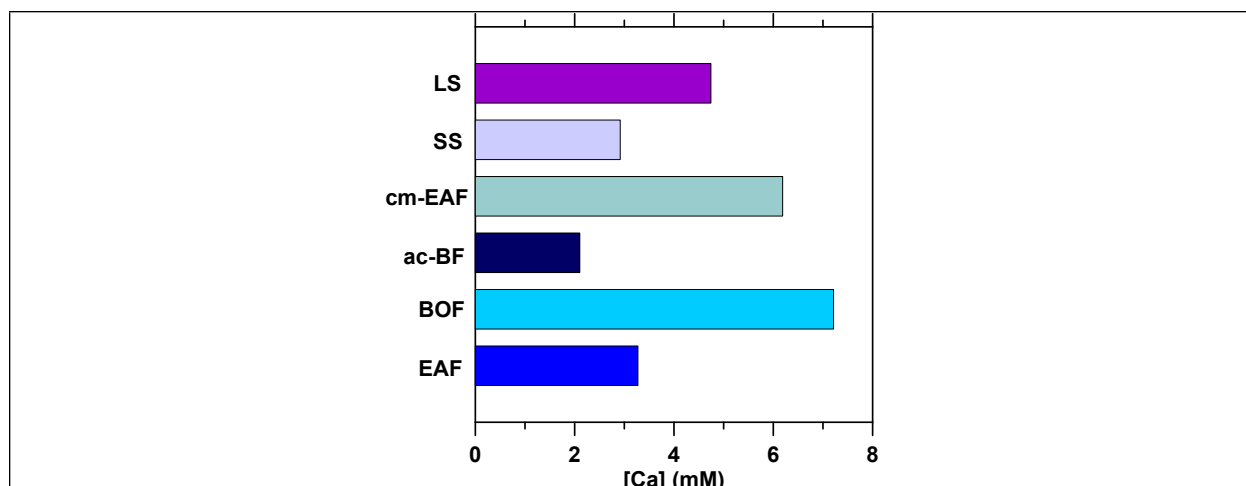


Figure S4. The Ca-concentration after 6 hours during leaching at 25 °C under stirred conditions ($s/l = 0.01$, particle size: 53 μm) for six different slag types including: ladle slag (LS), stainless steel slag (SS), co-mingled electric arc furnace steel slag (cm-EAF), air-cooled blast furnace slag (ac-BF), basic oxygen furnace slag (BOF), and electric arc furnace steel slag (EAF), respectively.

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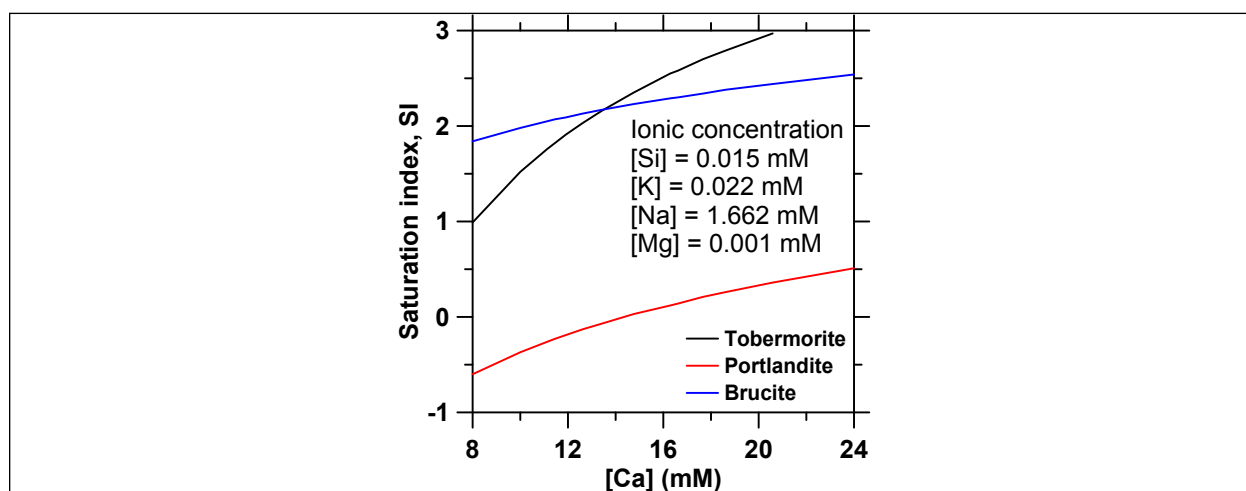


Figure S5. The saturation index of 11Å Tobermorite, Portlandite ($\text{Ca}(\text{OH})_2$) and Brucite ($\text{Mg}(\text{OH})_2$) as a function of $[\text{Ca}]$ and in the presence of other ionic species (see legend) as calculated using PHREEQC. The concentration of the other dissolved elements in solution was measured through ICP-OES for the 10 mM slag leachate solution after 6 h of leaching.

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15 **Ionic conductivity calculations**

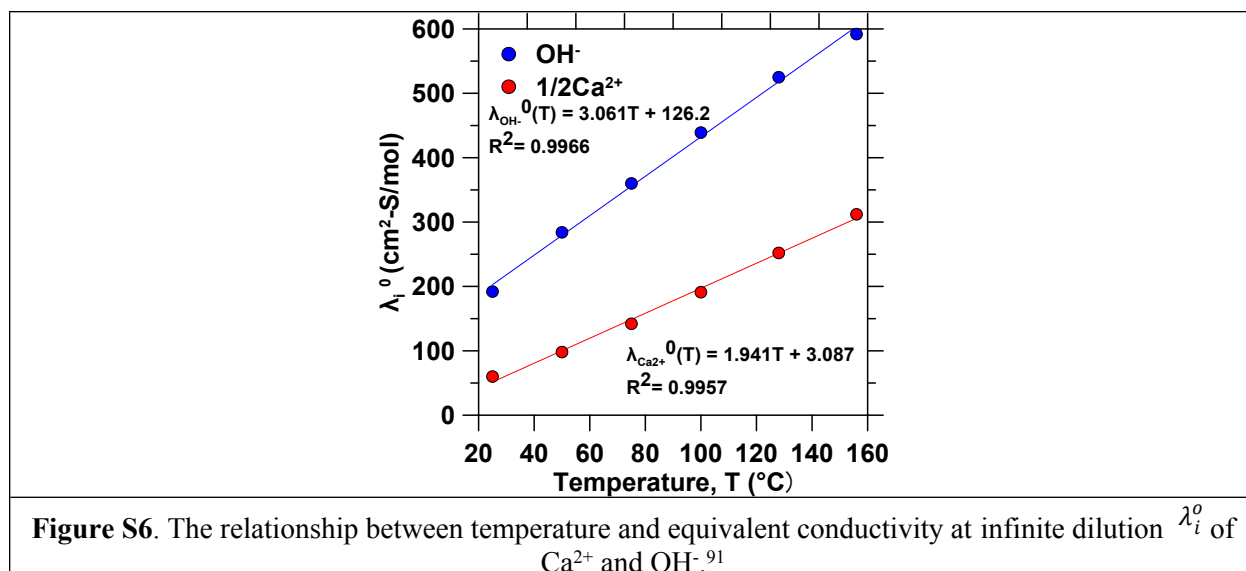
16 The parameters to solve Onsager's equation can be calculated as follows: The ionic conductivity
 17 of each species at infinite dilution λ_i^o is proportional to temperature in the range of our
 18 experiment. Figure S6 shows the reference value of λ_i^o at each temperature.⁹¹ Using the linear
 19 temperature dependence, λ_i^o is calculated as a function of temperature T.

20

$$\lambda_{Ca}^o(T) = 1.941T + 3.087 \quad (S1)$$

$$\lambda_{OH}^o(T) = 3.061T + 126.2 \quad (S2)$$

21



22

23 Moreover, S is calculated as

$$S_{Ca}(T) = \frac{1970 \times 10^6}{\{\varepsilon_r(T) \times T\}^{3/2}} \left(\frac{q(T)^*}{1 + \sqrt{q(T)^*}} \right) \times |z_{Ca} z_{OH}| \lambda_{Ca}^0(T) + \frac{28.98 \times |z_{Ca}|}{\eta(T) \times \{\varepsilon_r(T) \times T\}^{1/2}} \quad (\text{S3})$$

$$S_{OH}(T) = \frac{1970 \times 10^6}{\{\varepsilon_r(T) \times T\}^{3/2}} \left(\frac{q(T)^*}{1 + \sqrt{q(T)^*}} \right) \times |z_{Ca} z_{OH}| \lambda_{OH}^0(T) + \frac{28.98 \times |z_{OH}|}{\eta(T) \times \{\varepsilon_r(T) \times T\}^{1/2}} \quad (\text{S4})$$

24

25 where, T represents temperature, η the viscosity, ε_r the relative permittivity of the medium, and z_i
26 the valence of each ionic species. The parameter q can be calculated as

$$q(T)^* = \frac{2}{3} \times \frac{(\lambda_{Ca}^0(T) + \lambda_{OH}^0(T))}{(\lambda_{Ca}^0(T) + 2\lambda_{OH}^0(T))} \quad (\text{S5})$$

27

28 Moreover, the relative permittivity ε_r and the viscosity η of the solvent are dependent on
29 temperature T (in °C). Figure S7 shows the reference value of ε_r at each temperature.⁹²

30

31 The value of ε_r can be determined by the linear regression expressed as Equation S6 and η is
32 calculated by Equation S7.⁹³

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$$\varepsilon_r(T) = -0.3083T + 85.70 \quad (\text{S6})$$

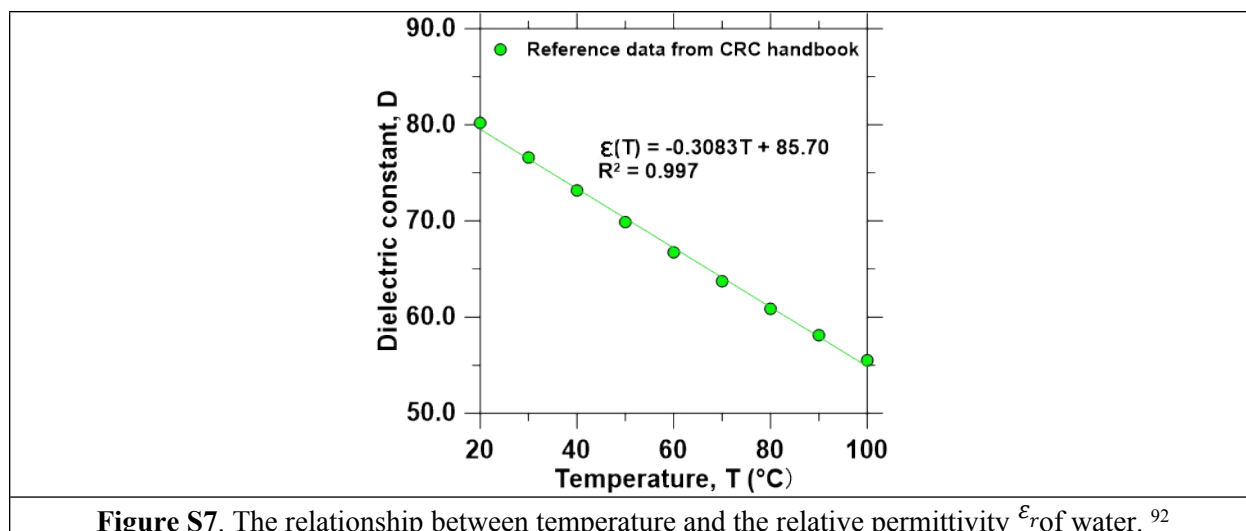
$$\eta(T) = 2.414 \times 10^{-5} \times 10^{247.8/(T-140)} \quad (\text{S7})$$

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35 Finally, the ionic concentration Γ can be calculated according to

$$\Gamma([Ca]_{aq}) = [Ca]_{aq} z_{Ca}^2 + [OH^-]_{aq} z_{OH}^2 \quad (\text{S8})$$

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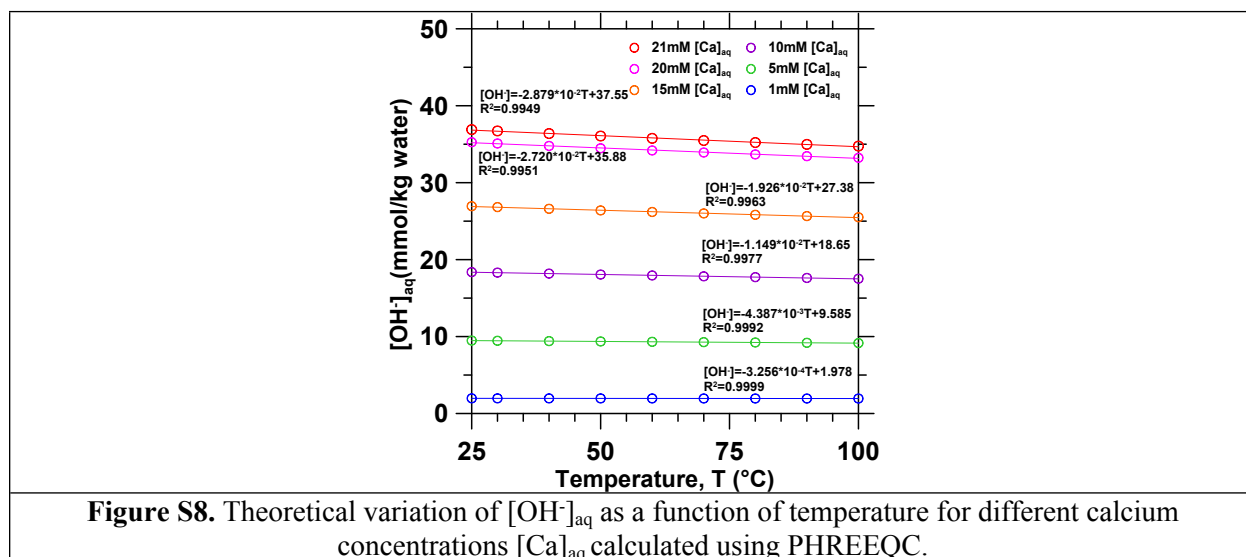


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38 **Determination of the calcium concentration from the conductivity of the solution**

39 To determine aqueous calcium concentration $[Ca]_{aq}$ from the measured conductivity, it is
 40 necessary to describe the relationship between the hydroxide concentration $[OH^-]_{aq}$, $[Ca]_{aq}$ and
 41 temperature because the concentration of hydroxide $[OH^-]_{aq}$ dominantly affects the conductivity.
 42 Figure S8 shows the theoretical variation of $[OH^-]_{aq}$ as a function of temperature for different
 43 calcium concentrations $[Ca]_{aq}$ calculated using PHREEQC. It indicates that $[OH^-]_{aq}$ decreased
 44 linearly with temperature.

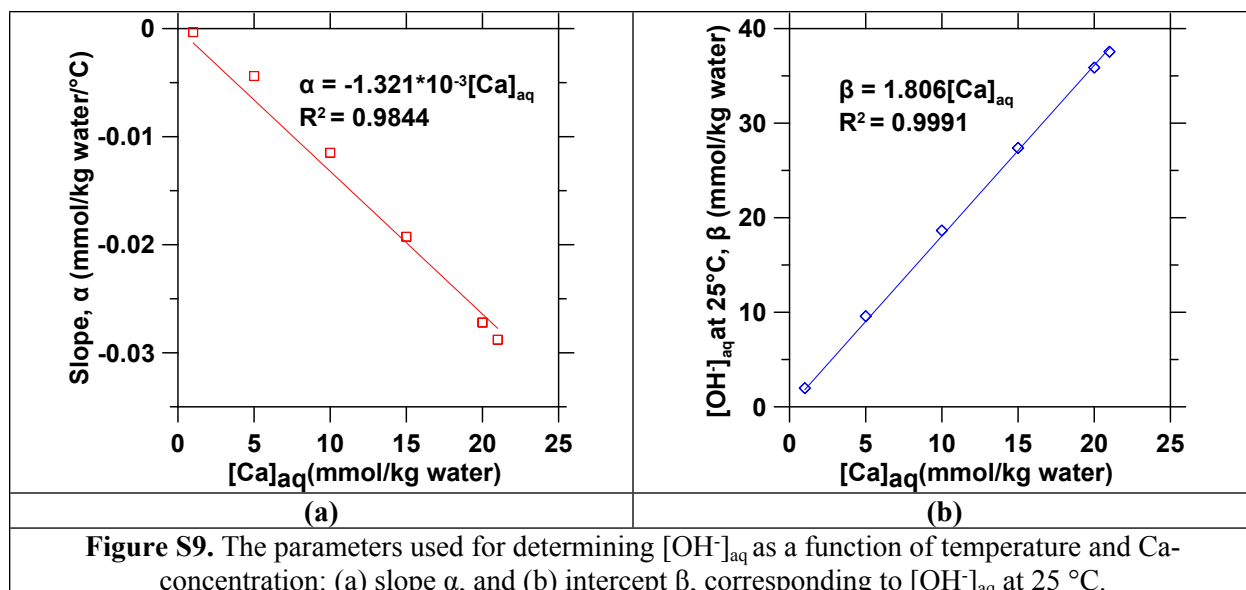
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47 The calibration equation was set as $[OH^-]_{aq} = \alpha T + \beta$, where β is $[OH^-]_{aq}$ at 25 °C as a function
 48 of $[Ca]_{aq}$ and α describes the change in $[OH^-]_{aq}$ with increasing temperature and $[Ca]_{aq}$. Figure S9
 49 shows the linear regression used to calculate α and β .

50



51

52 Hence, the change in $[\text{OH}^-]_{\text{aq}}$ as a function of temperature and calcium concentration is given by

$$[\text{OH}]_{\text{aq}} = -1.321 \times 10^{-3} [\text{Ca}]_{\text{aq}} T + 1.806 [\text{Ca}]_{\text{aq}} \quad (\text{S9})$$

53

54 Finally, Equation S10 can be used to estimate $[\text{Ca}]$ from conductivity measurements.

55

$$\sigma_{\text{calc}}([\text{Ca}]_{\text{aq}}, T) = 2[\text{Ca}]_{\text{aq}} \{ \lambda_{\text{Ca}}^{\circ}(T) - S_{\text{Ca}}(T) \times \Gamma([\text{Ca}]_{\text{aq}})^{1/2} \} + [\text{OH}]_{\text{aq}}([\text{Ca}]_{\text{aq}}, T) \quad (\text{S10})$$

56

57 Calculation of energy required to produce $\text{Ca}(\text{OH})_2$ using only waste heat

58 Concentrating the calcium-rich leachate solution to precipitate 1 kg of $\text{Ca}(\text{OH})_2$ using only waste
 59 heat would require the evaporation of 2466 L of water. First, the volume of water to be evaporated
 60 to bring the solution to the saturation point of $\text{Ca}(\text{OH})_2$ (13.8 mM at 100 °C) was calculated as
 61 $\Delta V = V_i - V_f$. Where V_i and V_f are the initial and final solution volume before and after evaporation,
 62 respectively. Assuming the leachate solution has an initial volume V_i of 5400 L and a $[\text{Ca}]$
 63 concentration c_i of 10 mM (Figure S1), the solution volume after evaporation V_f can be calculated
 64 according to $V_f = V_i c_i / c_f$, where $c_f = 13.8 \text{ mM}$ corresponds to the saturation concentration of
 65 $\text{Ca}(\text{OH})_2$ at 100 °C. Thus, 1487 L of water need to be evaporated. Second, precipitating 1 kg of
 66 $\text{Ca}(\text{OH})_2$ from this saturated solution requires to evaporate 979 L of water, assuming that for every
 67 liter of water evaporated from the saturated solution, 13.8 millimoles of $\text{Ca}(\text{OH})_2$ will precipitate.
 68 Thus, the total waste heat Q required to drive $\text{Ca}(\text{OH})_2$ concentration and precipitation using only
 69 waste heat is given by

$$Q = V \rho (c_p \Delta T + h_{fg}) \quad \text{S11}$$

70 where V (=2466 L) is the volume of water evaporated, $\rho = 1 \text{ kg/L}$ and $c_p = 4.18 \text{ kJ/(kg K)}$ are the
 71 density and heat capacity of water, respectively, $\Delta T = 75 \text{ K}$ is the temperature difference and
 72 $h_{fg} = 2257 \text{ kJ/kg}$ is the heat of vaporization of water. Under these assumptions, $Q = 6338 \text{ MJ}$ would
 73 be necessary to precipitate 1 kg of $\text{Ca}(\text{OH})_2$. By contrast, the total amount of energy for RO
 74 concentration and sub-boiling precipitation is 2.2 MJ of electricity and 677 MJ of waste heat,
 75 respectively. It is one order of magnitude lower than what is required to precipitate $\text{Ca}(\text{OH})_2$ using

76 waste heat only. Thus, the amount of waste heat of 5,000,000 GJ per year from a 10 MW_e mid-sized
77 power plant would enable the production of 7385 tons of Ca(OH)₂ per year when combined with
78 RO concentration. By contrast, the production rate would be only 788 tons of Ca(OH)₂ per year if
79 both concentration and precipitation are performed using only waste heat.