

SUPPLEMENTARY INFORMATION FOR:

***Development of a sample preparation procedure for Sr isotope analysis of
Portland cements***

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Abbreviations:

Sieving – sieving on 11- μ m sieve

KOSH - potassium hydroxide/sucrose solution

Conc. acid - Concentrated hydrochloric acid/nitric acid dissolution

Dil. acid - Dilute nitric acid dissolution

Equation:

$$\Delta_{abs} = \left| \left(\frac{{}^{87}\text{Sr}}{{}^{86}\text{Sr}} \right)_{\text{cement}} - \left(\frac{{}^{87}\text{Sr}}{{}^{86}\text{Sr}} \right)_{\text{clinker}} \right| \quad (1)$$

Legend:

Δ_{abs} - Absolute difference of processed cement from the clinker

$({}^{87}\text{Sr}/{}^{86}\text{Sr})_{\text{cement}}$ – Sr isotope ratio of processed cement

$({}^{87}\text{Sr}/{}^{86}\text{Sr})_{\text{clinker}}$ – Sr isotope ratio of the clinker

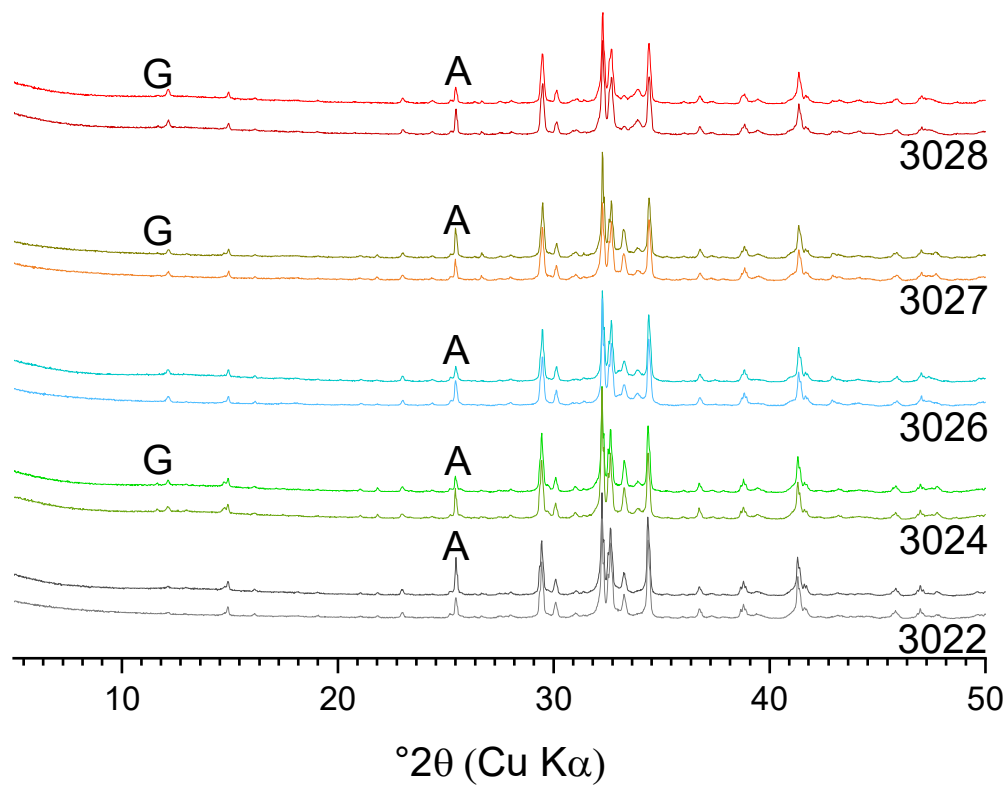


Fig. S1. X-ray diffractogram of non-treated cement (directly on top of the sample code) and sieved cement (on top of the respective non-treated pattern) for samples 3022, 3024, 3026, 3027 and 3028. G = Gypsum, A = Anhydrite.

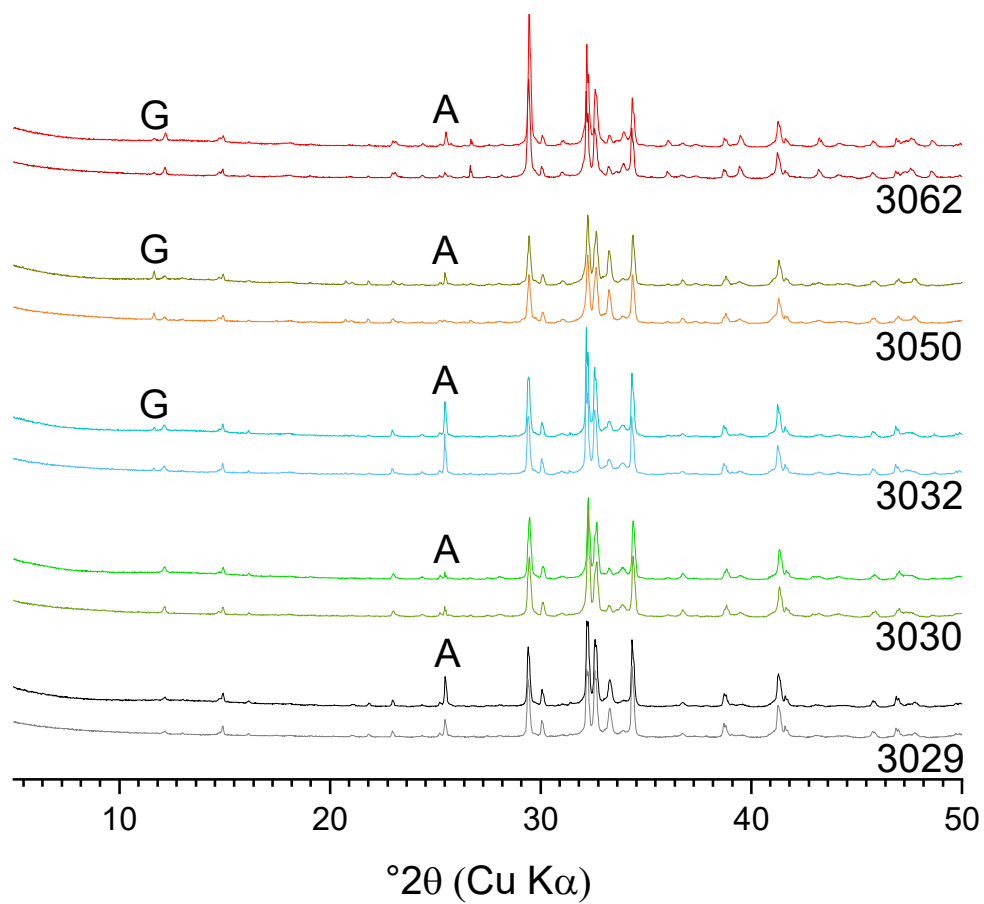


Fig. S2. X-ray diffractogram of non-treated cement (directly on top of the sample code) and sieved cement (on top of the respective non-treated pattern) for samples 3029, 3030, 3032, 3050 and 3062. G = Gypsum, A = Anhydrite.

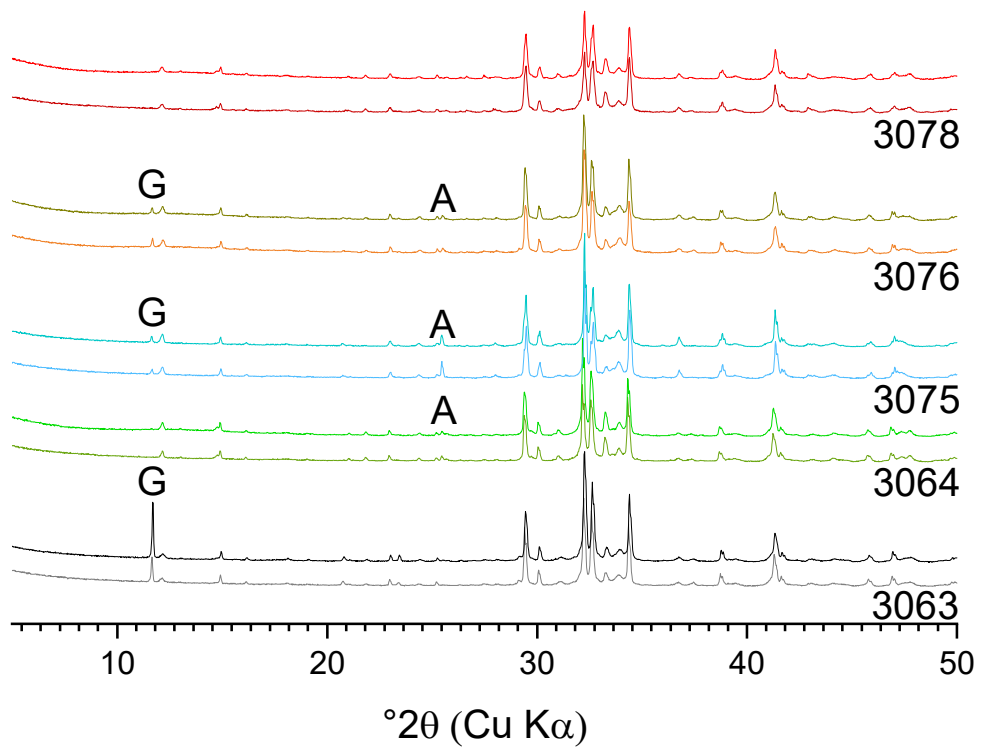


Fig. S3. X- ray diffractogram of non-treated cement (directly on top of the sample code) and sieved cement (on top of the respective non-treated pattern) for samples 3063, 3064, 3075, and 3078. G = Gypsum, A = Anhydrite.

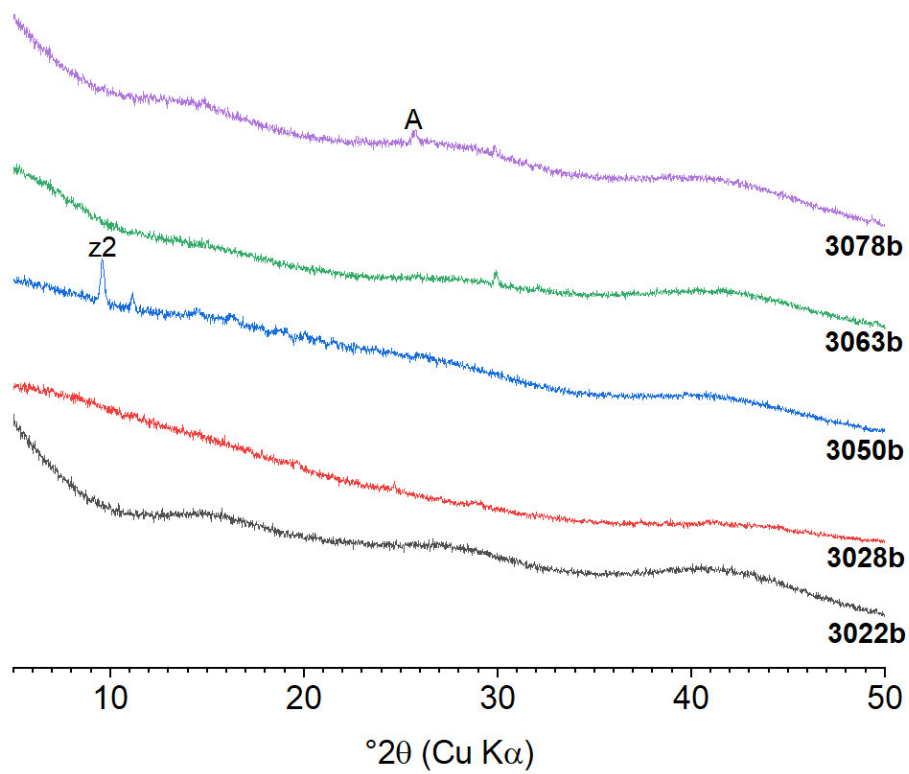


Fig. S4. X-ray diffractograms of the clinker samples 3022b, 3028b, 3050b, 3063b and 3078b after conc. acid treatment; A = anhydrite, z2 = zeolite-type phase.

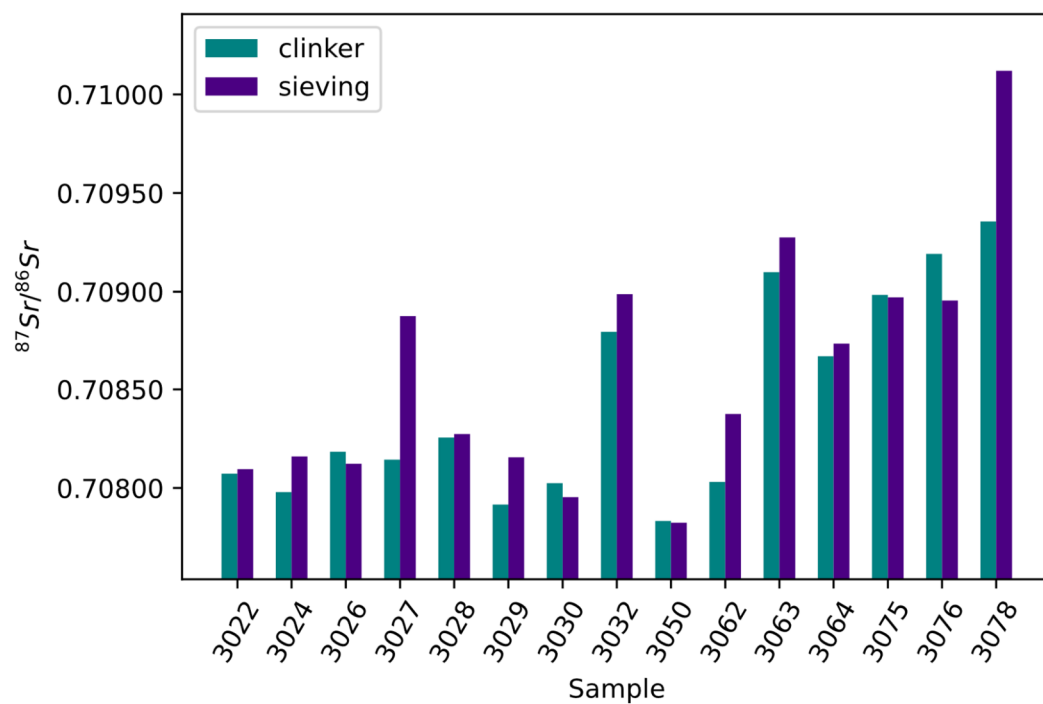


Fig. S5. Comparison of $^{87}\text{Sr}/^{86}\text{Sr}$ isotope ratios from fifteen cement samples prepared with sieving and subsequent conc. acid treatment (purple) and fifteen corresponding clinker samples after conc. acid treatment (blue-green).

Table S1. $^{87}\text{Sr}/^{86}\text{Sr}$ isotope ratio of processed clinkers, and cements after the KOSH and sieving treatment and clinker.¹ 2SE represents 2 x Standard Error (Standard Deviation divided by the square root of number of measured isotope ratios).

Sample	$^{87}\text{Sr}/^{86}\text{Sr}$ (clinker)	2SE x 10 ⁻⁵ (clinker)	$^{87}\text{Sr}/^{86}\text{Sr}$ (KOSH)	2SE x 10 ⁻⁵ (KOSH)	$^{87}\text{Sr}/^{86}\text{Sr}$ (Sieving)	2SE x 10 ⁻⁵ (Sieving)
3022	0.70807	1	0.70810	1	0.70807	1
3024	0.70798	1	0.70816	1	0.70811	1
3026	0.70818	3	0.70812	7	0.70809	1
3027	0.70814	1	0.70888	6	0.70886	1
3028	0.70826	1	0.70827	1	0.70825	2
3029	0.70792	1	0.70816	1	0.70785	1
3030	0.70802	1	0.70795	1	0.70794	1
3032	0.70880	1	0.70899	1	0.70858	1
3050	0.70783	1	0.70782	1	0.70783	1
3062	0.70803	3	0.70838	1	0.70824	1
3063	0.70910	1	0.70927	1	0.70918	1
3064	0.70867	1	0.70874	1	0.70849	1
3075	0.70898	1	0.70897	1	0.70863	1
3076	0.70919	1	0.70896	1	0.70880	1
3078	0.70936	1	0.71012	2	0.70951	1

¹ Each cement sample was loaded on a single filament, and the standard deviation was then calculated from minimum 100, maximum 200 measured isotope ratios.

Table S2. $^{87}\text{Sr}/^{86}\text{Sr}$ isotope ratio of processed cements after the dil. acid and conc. acid treatment.

Sample	$^{87}\text{Sr}/^{86}\text{Sr}$ (Dil. Acid)	2SE x 10^{-5} (Dil. Acid)	$^{87}\text{Sr}/^{86}\text{Sr}$ (Conc. Acid)	2SE x 10^{-5} (Conc. Acid)
3022	0.70804	1	0.70803	1
3024	0.70811	1	0.70812	1
3026	0.70808	1	0.70808	1
3027	0.70881	1	0.70881	3
3028	0.70823	1	0.70824	1
3029	0.70783	1	0.70786	1
3030	0.70793	1	0.70795	2
3032	0.70856	1	0.70859	1
3050	0.70781	1	0.70783	1
3062	0.70821	1	0.70821	1
3063	0.70917	1	0.70917	3
3064	0.70848	1	0.70848	1
3075	0.70860	1	0.70861	1
3076	0.70879	1	0.70881	1
3078	0.70946	1	0.70948	1

Table S3. Absolute difference Δ_{abs} between the $^{87}\text{Sr}/^{86}\text{Sr}$ isotope ratio of the cement and the $^{87}\text{Sr}/^{86}\text{Sr}$ isotope ratio of the corresponding clinker for the four investigated preparation methods. Δ_{abs} was calculated using equation (1).

Sample	Δ_{abs} (KOSH) $\times 10^{-5}$	Δ_{abs} (sieving) $\times 10^{-5}$	Δ_{abs} (dil. acid) $\times 10^{-5}$	Δ_{abs} (conc. acid) $\times 10^{-5}$
3022	2	0.03	4	4
3024	18	14	13	14
3026	6	10	11	10
3027	73	72	66	66
3028	2	1	3	2
3029	24	7	8	5
3030	7	8	9	7
3032	19	22	23	20
3050	1	0.3	2	1
3062	34	21	18	18
3063	18	8	7	7
3064	6	18	19	19
3075	1	36	39	37
3076	24	39	40	38
3078	76	16	10	12

Table S4. Results of the paired *t*-test for $^{87}\text{Sr}/^{86}\text{Sr}$ isotope ratios of clinker and KOSH $^{87}\text{Sr}/^{86}\text{Sr}$ isotope ratios of cements as calculated with Excel (Office 365).

t-Test: Paired Two Sample for Means		
	$^{87}\text{Sr}/^{86}\text{Sr}$ Clinker	$^{87}\text{Sr}/^{86}\text{Sr}$ KOSH
Mean	0.708436	0.708592
Variance	2.74E-07	3.8E-07
Observations	15	15
Pearson Correlation	0.891731	
Hypothesized Mean Difference	0	
df	14	
t Stat	-2.15852	
P(T<=t) one-tail	0.024369	
t Critical one-tail	1.76131	
P(T<=t) two-tail	0.048737	
t Critical two-tail	2.144787	

Table S5. Results of the paired *t*-test for clinker $^{87}\text{Sr}/^{86}\text{Sr}$ isotope ratios and sieving $^{87}\text{Sr}/^{86}\text{Sr}$ isotope ratios of cements as calculated with Excel (Office 365).

t-Test: Paired Two Sample for Means		
	$^{87}\text{Sr}/^{86}\text{Sr}$ Clinker	$^{87}\text{Sr}/^{86}\text{Sr}$ Sieved
Mean	0.708436	0.708429
Variance	2.74E-07	2.48E-07
Observations	15	15
Pearson Correlation	0.865751	
Hypothesized Mean Difference	0	
df	14	
t Stat	0.107146	
P(T<=t) one-tail	0.458097	
t Critical one-tail	1.76131	
P(T<=t) two-tail	0.916194	
t Critical two-tail	2.144787	

Table S6. Results of the paired *t*-test for clinker $^{87}\text{Sr}/^{86}\text{Sr}$ isotope ratios and dil. acid $^{87}\text{Sr}/^{86}\text{Sr}$ isotope ratios of cements as calculated with Excel (Office 365).

t-Test: Paired Two Sample for Means		
	<i>$^{87}\text{Sr}/^{86}\text{Sr}$ Clinker</i>	<i>$^{87}\text{Sr}/^{86}\text{Sr}$ Dil</i>
Mean	0.708436	0.708406
Variance	2.74E-07	2.41E-07
Observations	15	15
Pearson Correlation	0.873467	
Hypothesized Mean Difference	0	
df	14	
t Stat	0.447992	
P(T<=t) one-tail	0.330507	
t Critical one-tail	1.76131	
P(T<=t) two-tail	0.661014	
t Critical two-tail	2.144787	

Table S7. Results of the paired *t*-test for clinker $^{87}\text{Sr}/^{86}\text{Sr}$ isotope ratios and conc. acid $^{87}\text{Sr}/^{86}\text{Sr}$ isotope ratios of cements as calculated with Excel (Office 365).

t-Test: Paired Two Sample for Means		
	$^{87}\text{Sr}/^{86}\text{Sr}$ Clinker	$^{87}\text{Sr}/^{86}\text{Sr}$ conc
Mean	0.708436	0.708419
Variance	2.74E-07	2.41E-07
Observations	15	15
Pearson Correlation	0.877963	
Hypothesized Mean Difference	0	
df	14	
t Stat	0.259105	
P(T<=t) one-tail	0.399662	
t Critical one-tail	1.76131	
P(T<=t) two-tail	0.799325	
t Critical two-tail	2.144787	