

Supporting online material.

In situ disposal of crushed concrete waste as void backfill material at UK nuclear sites:

Leaching behaviour and effect of pH on element release.

By Danielle C. Tompkins¹, Douglas I. Stewart², James T. Graham³ and Ian T. Burke^{1*}

Consisting of:

2 Tables

5 Figures

2 Sections

SI Section A. Cement Chemistry Notation

CaO = C; SiO₂ = S; Al₂O₃ = A; Fe₂O₃ = F; H = H₂O (Neville, 2011)

Table S1. Mineral phases and chemical formulae referred to in this study. Cement chemistry notation is generic for C-S-H, C-A-S-H and AFm only, adapted from various authors (Matschei, Lothenbach and Glasser 2007; Blanc *et al.* 2010; Nonat 2004)

Notation	Mineral phase	Chemical formulae
C-S-H	Calcium silicate hydrate Tobermorite-like (Ca/Si ratio 0.66 – 0.83), jennite-like (Ca/Si ratio 1.25 – 1.50)	CaO-SiO ₂ -H ₂ O
C-A-S-H	Calcium silicate hydrate with alumina	CaO-Al ₂ O ₃ -SiO ₂ -H ₂ O
CH	Portlandite	Ca(OH) ₂
AFt	Ettringite	Ca ₆ Al ₂ (SO ₄)(OH) ₈
AFm	Aluminate ferrite monosulfate	Generic: 2CaO·Al ₂ O ₃ ·SiO ₃ ·12(H ₂ O)
Mc	Monocarboaluminate	Ca ₄ Al ₂ (CO ₃)(OH) ₁₂ ·5H ₂ O
Ms	Monosulfoaluminate	Ca ₄ Al ₂ (SO ₄)(OH) ₁₂ ·6H ₂ O
Hc	Hemicarboaluminate	Ca ₄ Al ₂ (CO ₃) _{0.5} (OH) ₁₃ ·5.5H ₂ O

Hyphens in C-S-H and C-A-S-H represent how the stoichiometry is variable (Richardson, 1999).

Table S2. Cement degradation stages for reference, adapted from Lagerblad (2005); Wieland (2014) and Jacques *et al.* (2010))

	Stage 1	Stage 2	Stage 3	Stage 4
pH	>12.5	~ 12.5	10.5 – 12.5	< 10.5
Dominant solution controlling phase	Na, K alkalis	Portlandite	C-S-H, AFm, AFt	Calcite,



Scale 1:800

Figure S1: Aerial photograph of four crushed concrete stockpiles (SP1-4) present on a UK Nuclear site. Each of four stockpiles are distinguished by different border colours. Yellow circles indicate the location of 21 trial pits where samples were recovered. Samples for this study were recovered from both surface and sub-surface layers from trial pits RS1 and RS4 on SP1.

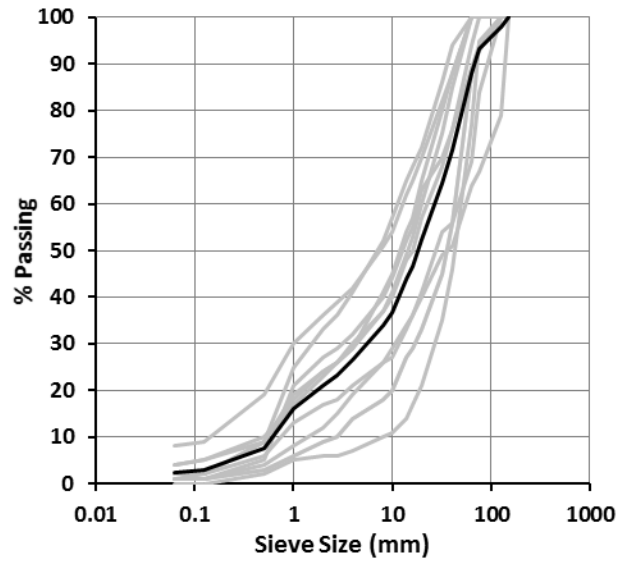


Figure S2. Particle size distribution curves of stockpiled concrete wastes collected from an average depth of 1.8 ± 0.6 m below surface level from ten trial pits on stockpile 1. Grey lines represent the individual samples, and the black line is mean of all samples.

S.I Section B: SEM method

Point counting analysis was completed by imposing 5 by 20 equally spaced lines on false-colour composite EDS images (example in SI Section 4; Figure S3). Phases were counted at the intersection between the horizontal and vertical lines. Only phases on the particle were counted and the totals were corrected where intersections were outside the particle such as in Figure S3. Five representative particles from the surface (0 – 10 cm) and the sub-surface (2.5 – 2.7 m) of the stockpile samples were used. An example is shown in Figure S3 below.

For simplicity, phases were either assigned as quartz (in blue), calcite (in red) or C-A-S-H (in varying shades of purple – from close to blue representing a Ca-depleted gel, to darker purple representing higher Ca/Si ratios). Where there were Fe, S or Mg particles (e.g. green particles) or Al-rich regions (lighter purples and blues in the C-A-S-H gel), this was also included under C-A-S-H. This is due to the inherent heterogeneity of C-A-S-H, and for ease of reference.

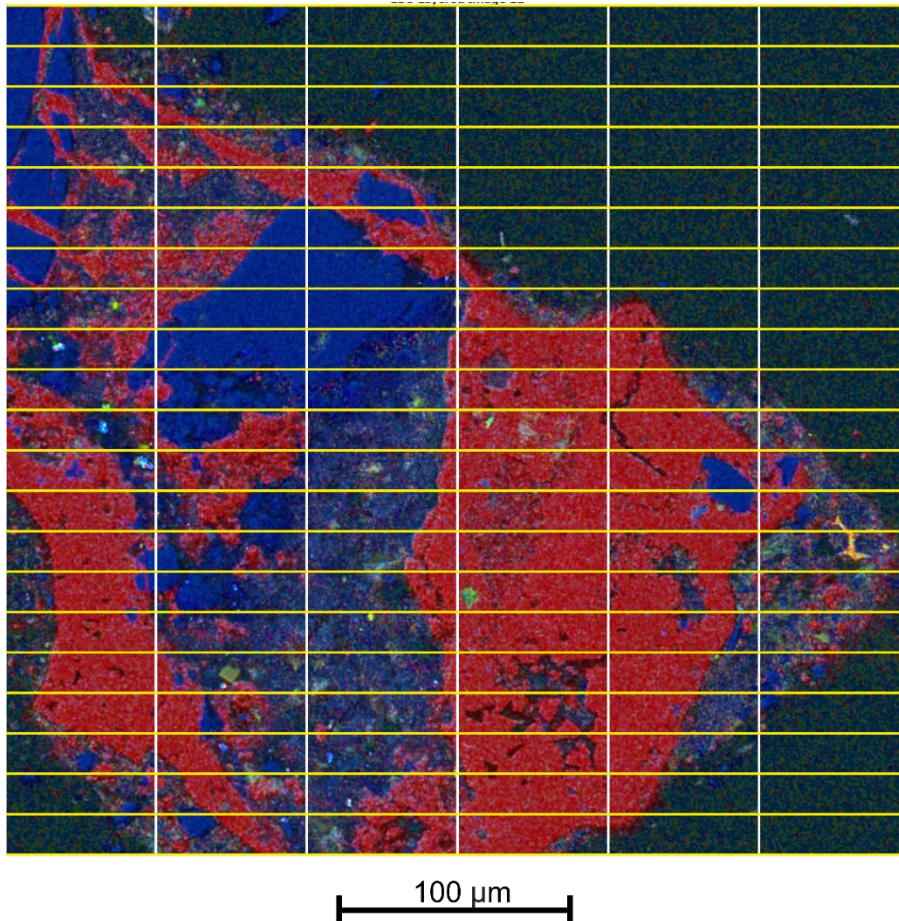


Figure S3: Example of grid used for point counting of false-colour composite EDS map of a sand-sized fraction from the surface 0 – 10 cm of stockpile 1 (trial pit RS4), where Blue = Si, Red = Ca, Green = Fe, Orange = Mg, Yellow = S, Gold = Al, Magenta = Na. Area outside the particle was not included in the totals which were corrected for.

S.I Section 4: Analysis and Results

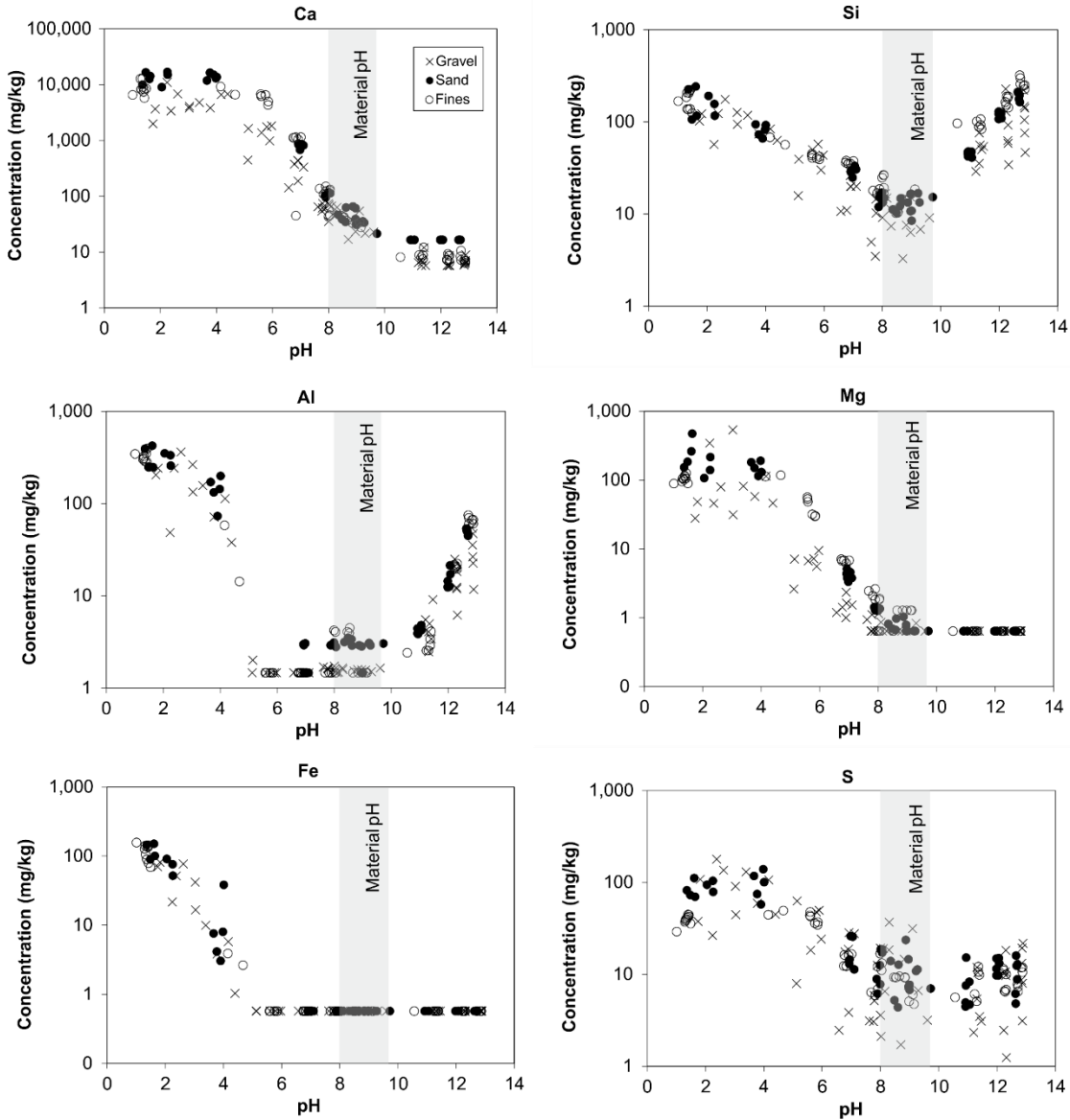


Figure S4. Leaching (mg/kg) of major elements Ca, Si, Al, Mg, Fe and S from the surface (0 – 10 cm) crushed concrete gravel, sand and fines fractions from trial pit RS1 and RS4 as a function of pH. Material pH denotes the range of aqueous pH values measured after suspension in deionised water for 7 days.

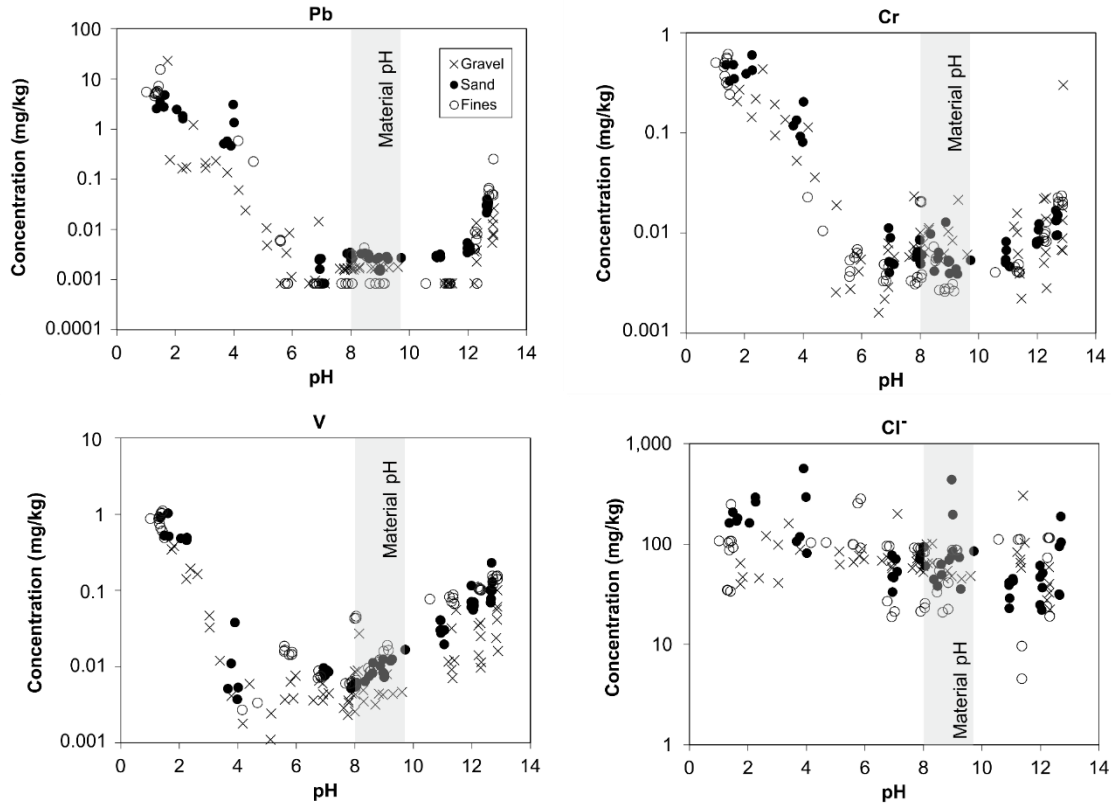


Figure S5. Leaching of Pb, Cr, V and Cl⁻ (mg/kg) from the surface (0 – 10 cm) crushed concrete gravel, sand and fines fractions from trial pit RS1 and RS4 as a function of pH. Material pH denotes the range of aqueous pH values measured after suspension in deionised water for 7 days.

References

- BLANC, P., X. BOURBON, A. LASSIN and E. C. GAUCHER. 2010. Chemical model for cement-based materials: Thermodynamic data assessment for phases other than C–S–H. *Cement and Concrete Research*, **40**(9), pp.1360-1374.
- JACQUES, D., L. WANG, E. MARTENS and D. MALLANTS. 2010. Modelling chemical degradation of concrete during leaching with rain and soil water types. *Cement and Concrete Research*, **40**(8), pp.1306-1313.
- LAGERBLAD, B. 2005. *Carbon dioxide uptake during concrete life cycle - State of the art*.
- MATSCHEI, T., B. LOTHENBACH and F. P. GLASSER. 2007. The AFm phase in Portland cement. *Cement and Concrete Research*, **37**(2), pp.118-130.
- NONAT, A. 2004. The structure and stoichiometry of C-S-H. *Cement and Concrete Research*, **34**(9), pp.1521-1528.
- WIELAND, E. 2014. *Sorption Data Base for the Cementitious Near Field of L/ILW and ILW Repositories for Provisional Safety Analyses for SGT-E2*. Nagra Technical Report 14-08.