

Recycled crushed concrete: engineering and environmental issues

Recyclage du béton concassé: aspects techniques et environnementales

S.A. Jefferis

Environmental Geotechnics Ltd, Oxfordshire, England

ABSTRACT: Much has been written about the sustainability benefits that flow from the use of recycled materials but little has been published about the risks associated with their use. This paper explores some of the issues that can arise from the use of recycled (crushed) concrete and in due course from its re-re-use or disposal when no longer required for use. It will be shown that its high pH leachate and susceptibility to sulfate attack can be of substantial concern.

RÉSUMÉ: Les avantages de l'utilisation de matériaux recyclés dans le cadre du développement durable sont bien connus mais les risques associés à leur utilisation n'ont pas été bien étudiés. Cette présentation étudie certains problèmes résultants de l'utilisation du béton recyclé, de sa réutilisation et de son élimination éventuelle. Deux problèmes importants seront considérés en particulier: le pH élevé du lixiviat et une attaque par des sulfates.

Keywords: Crushed concrete; recycled materials; re-re-use; pH; sulfates

1 INTRODUCTION

The construction industry is a major user of materials and so offers multiple opportunities for the re-use of materials particularly as it generates so much potentially re-usable material.

There is an extensive literature on the sustainability benefits that flow from the use of recycled materials and for some materials there is a substantial literature confirming that environmental impacts such as heavy metal concentrations in aqueous leachates are acceptable.

In contrast much less has been published about the long-term behaviour of recycled materials. Are they chemically and mechanically stable? Do they have potential for shrinkage, expansion or to damage structures formed from them or built upon them?

Furthermore, the environmental significance of more mundane chemical parameters such as pH tends to receive insufficient attention and there is inadequate precautionary advice.

This paper is focused on recycled crushed concrete as a material for use in construction. It is intended to provide a baseline from which to explore some of the problems that can arise from the re-use of construction materials. It will focus on the pH of waters which have been in contact with crushed concrete and more briefly on sulfate attack on crushed concrete.

2 USE, RE-USE AND RE-RE-USE

It is important to recognise that a fundamental aim of re-using materials is to keep them in the

cycle of utility and avoid their disposal to landfill. Use and re-use of construction materials therefore should be undertaken with full recognition that within a generation or two a structure may be demolished and that, at that time, its materials should be fit for return to the cycle of utility.

A use or re-use that dooms a material to landfill rather than a further re-use has bought time but potentially at the cost of very expensive landfill a generation later. Such action will be particularly unfortunate if the mass/volume of the material has been increased by blending it with virgin materials or with other recycled materials and the combined material later fails criteria for re-use or worse still, as has happened, results in the combined material being designated as not-inert and requiring expensive landfill. Similar problems can occur during use or re-use, if materials react with chemicals in adjacent materials and so become unfit for further re-use. The problems associated with multiple re-uses invites a thought: would an entropy based approach to recycled materials offer any insights?

3 USES FOR CRUSHED CONCRETE

Crushed concrete may be offered for very many purposes including:

- capping to protect subgrade and to provide a platform for further construction such as the sub-base for a road pavement or hard-standing;
- unbound subbase;
- temporary works such as working platforms;
- granular drainage media, for example, bedding or surround to drainage pipes – a situation particularly conducive to leaching by water;
- bulk fill.

It follows that in use, recycled crushed concrete will be in contact with natural soil and/or other construction materials and interaction with these materials should be considered when selecting recycled materials for use in construction. The potential for leaching by rainwater, surface

water, groundwater etc. also should be an important consideration.

4 REGULATORY CONTROLS

The leachate from crushed concrete and some other recycled materials can be strongly alkaline and it is therefore necessary to consider regulatory controls on pH. An upper limit of pH 9.0 is typically the control level set by the UK Environment Agency for discharges to controlled waters. Similar levels can be expected from other national environmental regulators.

As the pH of water flowing over crushed concrete can be much above pH 9, it has the potential to raise environmental concerns to the extent that treatment may be necessary before its release to the environment. As will be shown, this alkaline leachate may be generated for decades and so treatment also could be required for decades.

In contrast, if the alkaline leachate seeps into soil, it may pass unnoticed and/or be rapidly attenuated by interaction with the soil and especially fines/clays in the soil.

5 CHEMISTRY OF CONCRETE

To enable an assessment of the pH of leachate from crushed concrete it is first necessary to consider concrete production. Concrete is formed from a mixture of Portland cement, sand, coarser aggregates and water. The sand and aggregate, unless recycled materials themselves, tend to be relatively inert both physically and chemically. Indeed, inert behaviour is a key consideration when selecting aggregates for concrete production (though this is not always successful, consider alkali aggregate reaction) and noting that considerations of inert behaviour may not extend to the potential for environmentally toxic leachates in re-use situations.

In order to understand the chemical behaviour of crushed concrete it is necessary to consider the manufacturing process for Portland cements and

their principal components (Note: Portland cements are now designated CEM 1, CEM II etc. in the European Union. The term Portland cement will be retained for this paper as the CEM nomenclature is not universally used).

The raw materials for Portland cement production are a siliceous material such as clay or shale and a calcareous material such as limestone or chalk. These materials are fired in a kiln at a temperature of about 1500°C. After firing the clinker is ground and calcium sulfate is added to moderate the setting. At the kiln temperature, the raw materials combine to form four principal minerals, all oxide combinations:

- tricalcium silicate (C_3S) – reacts and hardens relatively rapidly on contact with water;
- dicalcium silicate (C_2S) – reacts and hardens slowly;
- tricalcium aluminate (C_3A) – hydrates rapidly and is responsible for the early age stiffening and setting (as distinct from hardening) of Portland cements. The early stiffening is moderated by the added calcium sulfate. It is this phase that is most sensitive to sulfate attack;
- tetracalcium aluminoferrite (C_4AF) – hydrates very slowly and contributes little to the hardened strength.

Note: for convenience, in cement chemistry oxides are represented by their first letter, thus C = CaO, S = SiO₂, A = Al₂O₃, F = Fe₂O₃.

Table 1 shows the typical oxide contents of three types of Portland cement and the calculated mineral composition for the cements.

On contact with water the silicate phases hydrate to form calcium silicate hydrates (CSH). These hydrates have an average calcium: silicon molar ratio of approximately 1.7 – lower than in the anhydrous minerals (Moir, 2003, Taylor, 1997). The excess calcium from the C_3S (molar ratio 3) and C_2S (molar ratio 2) is precipitated as calcium hydroxide. Thus the full hydration of one mole of C_3S will release about 1.3 moles of calcium hydroxide and one mole of C_2S on hydration will release 0.3 moles of calcium hydroxide.

The hydration reactions of C_3A and C_4AF are more complex and for the purposes of this paper it is sufficient to note that they can consume some of the calcium hydroxide released by the hydration of the calcium silicates.

Table 1. Typical composition of Portland cements (per cent by mass, data from Moir, 2003)

Cement type / Constituent	Grey	Sulfate resisting	White
CaO	64.8	64.3	69.1
SiO ₂	20.4	20.3	24.6
Al ₂ O ₃	5.1	3.6	1.9
Fe ₂ O ₃	2.9	5.1	0.3
MgO	1.3	2.1	0.55
Na ₂ O	0.11	0.10	0.14
K ₂ O	0.77	0.50	0.02
SO ₃	2.7	2.2	2.1
Calculated mineral compositions			
C_3S	57	62	67
C_2S	16	12	20
C_3A	9	0.9	4.5
C_4AF	9	16	0.9

Overall, on hydration, Portland cements may release some 15% calcium hydroxide by original dry mass of cement (N.B. it is difficult to determine the amount of calcium hydroxide in hydrated cement and the value determined is sensitive to the method employed).

Calcium hydroxide is strongly, alkaline; a saturated solution at 15°C has a pH of about 12.6. Thus hydrated Portland cement will be strongly alkaline. Portland cements also contain some sodium and potassium oxides (see Table 1). These will react with water to form strongly alkaline hydroxides so that fresh cement paste can have a pH over 13.5.

High pH is beneficial for reinforced and prestressed concrete as it inhibits the corrosion of embedded steel reinforcement and stressing cables surrounded by Portland cement grouts.

5.1 Leaching of concrete

For intact concrete exposed to above-ground environments, the high pH from calcium hydroxide has minimal environmental impact as the surface

concrete rapidly reacts with carbon dioxide (an acid gas) from the air to precipitate calcium carbonate. This tends to block the pores of the concrete slowing both further ingress of carbon dioxide and the leaching out of alkalis if water flows over the concrete. The rate of release of alkalis from intact carbonated concrete is usually sufficiently low that it passes unnoticed except sometimes at drips when calcium leached from concrete reacts with atmospheric carbon dioxide to form stalactites and, where the drips hit a lower surface, stalagmites. These are known as calthemites and will be predominantly calcium carbonate though there may be minor amounts of other minerals (for example, sulfate species).

Precipitated calcium carbonate can usefully clog or seal cracks, for example, in retaining walls which may develop a white crust on the exposed face.

Of course, more aggressive waters (e.g. higher bicarbonate content or acid content as can occur in some groundwaters) can damage concrete.

However, when concrete is crushed the reaction situation is very different. Crushing greatly increases the surface area of the concrete and internal cracks also will be created. The result is that if water flows over crushed concrete, it will leach alkalis and other chemicals at very much greater rates than from intact concrete. Reactions with external agents such as sulfates also will be accelerated.

6 CONTROLS ON THE RELEASE OF CHEMICALS

Before considering the effects of leaching, it is appropriate to consider the mechanisms that can influence and/or control the rate of release of chemicals from granular materials. These include:

- the diffusion rate of chemicals of concern through the solid phase. Low diffusion rates can be expected to lead to lower chemical concentrations in leachates. If diffusion is the principal

control, leaching will be more rapid from smaller particles;

- surface processes controlling dissolution at the solid-liquid interface (for example, dissolution may follow simple first order reaction kinetics or more complex higher order kinetics);
- the leach water flow rate, that is the water supply rate. Low / high flow rates can be expected to give high / low chemical concentrations in the leachate – if there are also diffusion and/or surface process controls;
- the solubility of the relevant chemical species.

In many practical situations for coarse crushed concrete, diffusion may be the principal control on leaching. Water supply rate may control leaching for fine material or if the crushed concrete is kept damp (e.g. beneath a slab) and subject to leaching only by seasonal wetting and drying so that there is diffusion throughout the year with leaching predominantly in wet seasons.

It should be noted that the above analysis has considered only leaching. There will be other processes and chemical reactions such as sulfate attack which is briefly discussed at Section 11.

7 ALKALINE MATERIALS IN HYDRATED PORTLAND CEMENT

The leaching of alkalis from Portland cements is complex as there are a number of alkaline species in the cements. The principal sources of alkalis in hydrated Portland cements are set out below.

7.1 *Sodium and potassium hydroxides*

On wetting Portland cements, sodium and potassium hydroxides are formed from the hydration of sodium and potassium oxides present in the original dry cement powder. Both hydroxides are infinitely soluble in water and readily leached. The principal control on their leaching will be the availability of water and their rate of release from the crushed concrete. Sodium and potassium hydroxides can take leachate pHs to over 13.0.

7.2 Calcium hydroxide

As previously discussed, calcium hydroxide (hydrated lime, $\text{Ca}(\text{OH})_2$) is produced by the hydration of the calcium silicates in Portland cements. Calcium hydroxide is a solid of low solubility (about 1.7 g/litre – the solubility reduces with temperature) and as a result its rate of leaching can be limited by solubility constraints as well as the availability of water and its release rate from the crushed concrete.

7.3 Decalcification of calcium silicate hydrates

Calcium silicate hydrates are the principal reaction products that give strength to hardened Portland cements. These show an alkaline reaction. If they are leached by water so that the pH drops below that of saturated calcium hydroxide, calcium hydroxide will be released from the calcium silicate hydrates, so lowering the calcium to silicon ratio in the remaining hydrates.

Leaching from calcium silicate hydrates is likely to be slower than from free calcium hydroxide and water flow rate will become a progressively more important control on the leaching rate as the pH drops towards 10.5 (reduction in pH indicating a reduction in the hydroxyl ion concentration in the water and so also the calcium ion concentration).

7.4 Dissolution of remnant calcium silicate hydrates

Continued leaching will reduce the calcium to silicon ratio of the calcium silicate hydrates and the pH also will drop. When the pH reaches about 10.5, calcium and silicon dissolve congruently from the calcium silicate hydrates so that they dissolve in their entirety. This final stage will take a very substantial time as the dissolved concentrations will be low. Dissolution of the hydrates will lead to a significant loss of strength of the crushed concrete.

8 LEACHING CRUSHED CONCRETE

In order to make any quantitative estimate of the timescales for leaching it is necessary to know the original mix parameters. However, the Portland cement content of any concrete will be a function of many variables and especially the use for which it was prepared. Those undertaking the crushing and those using the crushed material are most unlikely to have any knowledge of the original mix design including factors that would have been important for its original use such as water/cement ratio – a key factor for the durability of the intact concrete which will, in turn, influence the behaviour of its crushed counterpart.

The re-user also will not know what cement type was used nor whether additions (fly ash, ground granulated blastfurnace slag, limestone powder etc.) were used – again these will influence the behaviour of the crushed material.

Furthermore, the crushed concrete may not be of a single concrete type or come from a single source. It may be a mix of concrete types/sources.

As an example mix for this paper, a crushed concrete prepared from a concrete with a cement content of 350 kg/m^3 is assumed. For its re-use, it is further assumed to have been compacted to a porosity of 20%. Key parameters for the resulting crushed concrete are shown in Table 2, including possible calcium oxide contents calculated from the data shown in Table 1

9 LEACHING VOLUME AND TIME

As discussed at Sections 7.3 and 7.4, the leachate from calcium silicate hydrates can have a pH at or above 10.5 until all the CSH has been dissolved. Thus at all stages of leaching, the pH can be greater than the typical regulatory control value, pH 9.0 unless the release rate from the crushed concrete is low relative to the water flow rate so that this pH is not reached and/or the pH is reduced by chemical reactions.

For the example mix shown in Table 2, indicative volumes of water to leach calcium oxide

from the concrete have been estimated for two leaching stages with the following assumptions:

(a) initial leaching as calcium hydroxide at 1.7 g/litre, mass of calcium hydroxide leached 10% of initial dry mass of cement.

(b) final congruent dissolution of the CSH assumed to occur once the C:S molar ratio has reached 0.85, pH 10.5. The concentration of calcium estimated as 65 mg/litre in this cementitious environment.

Table 2. Assumed concrete parameters – an example

Parameter	Value	Units
Example cement content	350	kg/m ³
Assumed porosity of compacted crushed concrete	20	%
Mass per unit volume of cement in compacted crushed concrete	280	kg/m ³
Calcium oxide content of cement	64.8	%
Mass of calcium oxide in compacted crushed concrete	181	kg/m ³
(a) Volume of water to leach 10% calcium hydroxide by mass of cement, pH about 12.6	16.5	m ³ /m ³
(b) Volume of water for congruent dissolution of CSH at C:S ratio 0.85, pH about 10.5	500	m ³ /m ³
Retardation (a)	83	-
Retardation (b)	2500	-
Example leaching rate	3	m ³ /m ³
(a) Time to leach 10% calcium hydroxide by mass of cement	5.5	years
(b) Time for congruent dissolution of CSH	165	years

Of course, there will be intermediate stage(s) as the pH drops from 12.6 to 10.5 (see Section 7.3). This might require perhaps 70% of the volume of water for Stage (b).

From Table 2, it can be seen that the estimated leach water volume for Stage (a) is 16.5 m³ water per m³ crushed concrete. For the final stage (b), congruent dissolution, the volume is about 500 m³/m³. Fuller analyses can be made but these

would require more data on the chemistry of the crushed concrete and the leaching water.

A perspective on these volumes can be obtained by considering a rainfall rate. If it is assumed, by way of example, that the leaching water is applied at 1.5 m³/m²/year over 0.5 m depth of crushed concrete (as rainwater but ignoring carbonic acid and any other acids in the rainwater) then the timescale over which leachates may be strongly/significantly alkaline is from 5.5 to 165 years (the latter figure is of course very reassuring for those concerned with the durability of concrete leached by water).

The above indicative volumes and times assume equilibrium has been achieved between the leaching water and the cement hydrates and thus that the stated pHs have been reached. They are therefore minimum volumes and times. In practice equilibrium may not be achieved and volumes will be larger, times longer and pHs lower. Despite this in practical situations the inventory of alkalis in crushed concretes is likely to be so large that the pH of any water running through it can be strongly alkaline and above regulatory control limits for decades.

It should be noted that during early leaching whilst sodium and potassium hydroxides remain in the crushed concrete, the pH may be >12.6.

10 RETARDATION

Table 2 shows that the volumes of water for the two leaching processes considered (a, b) are very different. It follows that if there is continuing flow of water through crushed concrete the various interaction processes will move through it at very different rates. The rate of advance of a process can be related to the volume of water per cubic metre of compacted crushed concrete N required to drive it forward. The velocity u of the reaction front relative to the interstitial water velocity v (flow rate per unit cross sectional area/porosity e of the crushed concrete) is:

$$u = \frac{v}{1+N/e} \quad (1)$$

The factor $(1 + N/e)$ is the retardation. Retardations have been included in Table 2 from which it is clear that the processes described in Sections 7.1 to 7.4 will move through water leached concrete at very different rates.

10.1 Neutralisation reactions

The above calculations have ignored other processes and reactions that can affect the pH of leachates. For example, rainwaters contain alkali reactive species that can reduce leachate pH. These species include acids from the solution of carbon dioxide and sulfur and nitrogen oxides (SO_x and NO_x) from air. Below ground, the equivalent carbon dioxide concentration in groundwater can be many times that in equilibrium with the carbon dioxide in air.

The reactions of both dissolved carbon dioxide species and acid rain species with crushed concrete are likely to be rapid and will occur at or near the location of first contact of the water and crushed concrete.

Water in equilibrium with atmospheric carbon dioxide will have a pH of about 5.6. Its alkali neutralisation front will propagate very slowly with an initial retardation factor estimated to be $>10^5$ for the crushed concrete shown in Table 2 (though this will drop as acid reactive species are leached). It follows that until a significant fraction of acid reactive species has been removed, the neutralisation front will be more retarded than the fronts of the leaching processes. Neutralisation will reduce the amount of alkali in the crushed concrete near the first contact of water and crushed concrete. However, if there is a significant volume of crushed concrete, the water, now with acid species neutralised, will flow on tending to develop its full un-neutralised pH during contact with the remaining crushed concrete.

There also will be neutralisation of the alkalis in crushed concrete by atmospheric carbon dioxide if air moves through it as a result of atmospheric pressure changes and/or by diffusion.

Achieving a significant degree of neutralisation may take a substantial time.

11 AGGRESSIVE CHEMICALS

Intact concrete can resist aggressive chemicals such as sulfates for a considerable time because of the time it takes for chemicals to penetrate. However, crushing greatly increases the surface area to volume ratio and this together with crushing induced cracks can lead to much more rapid chemical reactions with the potential for the loss of materials performance and structural integrity. Unfortunately published research on this is minimal – a matter which needs to be addressed.

Sulfate attack should be considered if sulfur containing species including sulfates and/or sulfides are present in soils adjacent to the crushed concrete or in material placed with it. Attack can be both rapid and severe with the formation of both ettringite and thaumasite (note: thaumasite formation requires a source of carbonate such as limestone aggregate) leading to substantial expansion and disintegration of the crushed concrete.

It follows that guidance for intact concrete such as BRE Special Digest 1 (BRE, 2005) must not be used for crushed concrete. Crucially, BRE (2011) states recycled crushed concrete aggregate (RCA): “should not be used where it is subject to conditions equivalent to aggressive ground category ACEC Class 2 or above. For neutral pH conditions (ie no acidity) this equates to not using RCA when sulfate levels are Class DS-2 or above. The lower limit of Class DS-2 is 400 mg/l SO_4 for sulfate in groundwater, and 500 mg/l SO_4 for sulfate in a 2:1 water:soil extract test.” (Note: ACEC: Aggressive chemical environment; DS: Design sulfate).

12 OTHER RISKS

Risk of clogging: the alkaline calcium containing leachates from crushed concrete will precipitate

calcium carbonate on contact with air or with waters containing inorganic carbon species. There is therefore a risk of blocking drainage systems etc.

Risks from associated materials: as crushed concrete will come from the demolition of buildings and other structures, there is a risk that it may contain unwanted and potentially undesirable materials, such as asbestos from asbestos insulation, asbestos cement sheeting and pipes. Fibres may be released by the crushing processes. The potential for harmful materials should be included in risk assessments for all recycled materials; risks must be appropriately managed to protect all those involved from the production process, through storage, transport, re-use, future demolition and re-re-use and all associated laboratory testing

Risks from other chemically reactive materials offered for re-use in construction: These may give rise to potentially hazardous high pH leachates and deleterious expansion. These include the "secondary" materials: steel slag, old blast furnace slag and incinerator bottom ash aggregate (IBAA) derived from the incineration of municipal solid waste. Problems associated with the ferrous slags are discussed in BRE IP/05 (BRE, 2005). Several expansive reactions can occur in IBAA, notably reaction of metallic aluminium in the IBAA with water in the alkaline environment of the ash, and then further reactions of aluminium reaction products (Eden, 2017).

13 CONCLUSIONS

Crushing concrete greatly increases its surface area to volume ratio and even relatively intact pieces of crushed material may have a network of internal cracks. As a result crushed concrete is much more rapidly reactive than intact concrete. Two issues, pH and sulfate attack have been examined but others may need to be considered when using this material. The key messages of this paper are therefore:

- be aware that recycled materials introduce a new spectrum of potential hazards. They are not

proven materials in the sense that regular natural aggregates are. In many circumstances they may behave in a benign manner but problems have occurred and will continue to do so without more research and the development of focused guidance documents;

- testing recycled materials for environmental acceptability alone is not sufficient. Recycled materials must be tested for in-service behaviour in in-service environments;
- think, this use, next use, future uses. Do not let an incautious re-use deny future re-uses.

14 ACKNOWLEDGEMENTS

The author gratefully acknowledges his many fruitful discussions on construction materials with Mr Ian Longworth formerly of the Building Research Establishment, Dr Adrian Bath of Intelisci and Mr Michael Eden of Sandberg Consulting Engineers. Their advice over many years has been invaluable. However, the author remains responsible for the opinions and analyses presented in this paper.

15 REFERENCES

- Building Research Establishment (BRE) 2005. *Special Digest 1 Concrete in aggressive ground*, 3rd edn, Watford.
- Building Research Establishment (BRE) 2011. *DG 522. Hardcore for supporting ground floors of buildings: Part 1. Selecting and specifying materials*, Watford.
- Dunster, A.M., Longworth, T.I., Nixon, P.J., Lane, S.J. and Collins, R.J. 2005. *BRE Information Paper IP/05. Development of sites containing expansive ferrous slags*, Watford.
- Eden, M. 2017. Mechanisms of expansion in incinerator bottom aggregate (IBAA) used as aggregate in concrete and as fill in engineering applications. *Proceedings of the 16th Euroseminar on Microscopy Applied to*

Building Materials, Les Diablerets, Switzerland.

Moir, G. 2003. *Cements*, in Newman, J. and Choo, B.S. *Advanced concrete technology, Constituent materials*. Butterworth Heine-
man, Oxford.

Taylor, H.F.W. 1997. *Cement chemistry*, 2nd edn. Thomas Telford, London.