RELEASE OF DANGEROUS SUBSTANCES FROM CONSTRUCTION AND DEMOLITION RECYCLED MATERIALS USED IN ROAD PAVEMENTS - LABORATORY AND FIELD LEACHING TESTS[1]

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Abstract

The use of Construction and Demolition Recycled Materials in road pavements is envisaged as a recycling solution with environmental and economic benefits, allowing the incorporation of large amounts of those materials from different sources. The environmental concerns regarding the potential contamination of surface and ground waters and soil forces the evaluation of the leaching behavior of the recycled materials. In Portugal the application of Construction and Demolition Waste (CDW) in civil engineering works demands their compliance with the requirements of leachability for waste admissible in landfills for inert waste. Nevertheless some issues concerning this criterion and the leaching procedure should be better studied whereby leaching tests, laboratorial batch and column leaching tests and *in situ* lysimeter leaching tests, were performed, or are still ongoing, on recycled aggregates – crushed mixed concrete, crushed reclaimed asphalt and milled reclaimed asphalt – and on a natural aggregate to provide a comparison with the results obtained with recycled materials. Crushed and milled reclaimed asphalt aggregates do not fulfill the criteria to be disposed at landfill for inert waste owing to the content of the hydrocarbons C10-C40. Nevertheless, based only on the thresholds for leachability defined in the Portuguese technical specifications no restrictions apply for their use from an environmental perspective.

1. INTRODUCTION

In the European Union the overall annual material consumption per inhabitant is around 16 tonnes with almost one third being wasted and about half of this being landfilled, thereby hindering a closed loop material cycle. One of the aims of the Europe 2020 strategy and also an opportunity according to the Vision 2050 of the World Business Council for Sustainable Development is the efficient use of resources. This concern is further reinforced in the Construction Products Regulation (CPR) No. 305/2011 [1] which points out the need for recyclability of materials from construction and demolition works and the use of raw materials and secondary materials with low environmental impact.

The aggregates sector, with a world estimated annual demand of 5.2% is the non-energy extractive industry using more natural resources and can be seen as a priority sector to improve resource efficiency by increasing the use of recycled aggregates, i.e. materials from CDW that, currently in Europe, represents only 5% of the aggregates production. Nevertheless, to boost the use of recycled aggregates the materials from CDW stream should be considered secondary raw materials whereby they must obtain the end of waste (EoW) status under the Waste Framework Directive [2].

According to EoW criteria one important condition is that the materials that cease to be waste must not lead to overall adverse environmental or human health impacts, an issue also recognised in the basic requirement of CPR. To provide evidence on this topic the leachability of recycled aggregates must be addressed and compared to threshold values for the relevant pollutants. Development of a methodology to establish limit values for pollutants in the EoW framework at European level is ongoing but in its lack each Member State defines their own criteria [3]. Similarly to other countries, Portugal implemented in their technical specifications for the use of recycled CDW aggregates a leachability requirement. According to this requisite the eluates of recycled aggregates, obtained using the batch leaching test of EN 12457-4 [4] should comply with the limits for waste to be disposed in landfill for inert waste.

Recycled aggregates are increasingly common in road infrastructures although the use of crushed or milled reclaimed asphalt (RAP) as unbound materials for base and subbase layers is unusual. The National Laboratory for Civil Engineering (LNEC) and the University of Lisbon (IST) carried out the research project, SUPREMA, regarding the use of recycled aggregates in road infrastructures including those materials [5, 6]. To assess their potential release of pollutants, the results from leaching tests were evaluated against the leaching behaviour of an often used recycled aggregate - crushed mixed concrete – and of a natural aggregate – limestone – for the abovementioned application.

The European standard regarding the use of unbound and hydraulically bound materials in road construction foresees a laboratory batch leaching test, according to EN 1744-3 [7]. In theory, leaching according to EN 12457-4 [4] and EN 1744-3 [7] evaluate the short term release of contaminants based on the principle that equilibrium or near equilibrium conditions between solid and liquid phases were achieved, but some debate around this subject can be found in the literature [8]. The repeatability of the test is uncertain owing to the extended particle size allowed and there is a mix of release mechanisms.

Moreover, the release of pollutants was also evaluated using lysimeters in which a layer of each recycled aggregate, placed under a drainage layer, was compacted with the optimum water content of the Modified Proctor test. The tests conditions of the lysimeters simulate the field conditions of the road test sections constructed during the project but no wearing course was placed above the recycled aggregates, corresponding to the extreme situation of a damaged pavement due to traffic and climatic solicitations, whereby it would result in higher leachability.

The main objective of this paper is to present and analyse the leachability observed in different leaching tests, assuming that in all situations the recycled aggregates were tested under similar conditions.

2. MATERIALS AND METHODS

The leaching tests performed and the constituents of the crushed mixed concrete, CMC, crushed reclaimed asphalt, CRAP, and milled reclaimed asphalt, MRAP are shown in Table 1 and in Table 2, respectively.

Often the leaching of heavy metals and salts are considered the main categories of concern regarding the use of recycled aggregates. Nevertheless, this study was mainly focused in RAP aggregates whereby an analysis of organic analytes was also carried out.

Table 1: Test methods for assessment of leaching

EN 1744-3	Preparation of eluates by leaching of aggregates
EN 12457-4	Characterization of waste. Leaching. Compliance test for leaching of granular waste materials and sludges – Part 4: One stage batch test at a liquid to solid ratio of 10l/kg for materials with particle sizes below 10mm (without or with size reduction)
In situ	Recovery and analysis of leachates from lysimeters percolated by rainwater, with cumulative L/S ratio from 0.1 to 10.0.

Table 2: Proportion of the constituents in the recycled aggregates [9]

Constituents		CMC	CRAP	MRAP
Rc [%]	Concrete, concrete products, mortars	60	6.1	0.0
Ru [%]	Unbound aggregate, natural stone, hydraulically bound aggregates	24	29	0.0
Ra [%]	Bituminous materials	12	64	99
Rb [%]	Clay masonry units (i.e. bricks and tiles), calcium silicate masonry units, aerated non-floating concrete	3.9	0.9	0.0
Rs [%]	Soils	0.0	0.0	0.0
Rg [%]	Glass	0.0	0.0	0.0
X [%]	Other: cohesive materials (i.e. clay and soil), plastics, rubbers, metals (ferrous and nonferrous), non-floating wood, gypsum plaster	0.1	0.0	1.0
FL [cm^3/g]	Floating particles	0.0	0.0	0.0

3. RESULTS

3.1 Batch leaching tests

Table 3 shows the results of leaching tests performed according to EN 12457-4 [4] as well as the allowed limits of leachability for recycled aggregates to be used in civil engineering works, as foreseen in the Portuguese specifications for these materials. As can be seen the released contents of substances of concern are too far from those limits.

In what concerns to eluates from RAP aggregates, the dissolved organic carbon, especially for MRAP, was higher when compared to CMC owing to the high content of the bituminous binder (see Table 2) tough it is lower that the limit for landfills for inert waste. Nevertheless, an increased mobility of some determinands due to the higher content of dissolved organic carbon in RAP aggregates, as seen in eluates of other residues [10], was impossible to identify in this leaching test.

If all the requirements for inert waste, stated in the Landfill Waste Framework, were taken into account to allow the use of recycled aggregates it is also necessary to analyse the total content of organic contaminants. RAP aggregates show a high total organic carbon content, particularly MRAP, that is not a problem since the content of dissolved organic carbon in the eluate is below the threshold for acceptance of waste at landfills for inert waste. Other organic contaminants – PAH, BTEX and PCB – show lower contents on RAP aggregates but for hydrocarbons C10-C40 the threshold of 500 mg/kg is exceeded: the contents were 6200 mg/kg for MRAP and 3020 mg/kg for CRAP. Hence, RAP aggregates will not be classified as inert and could not be used as unbound aggregates.

As regards the inorganic metal analytes, chromium shows the higher content in CMC eluates tough it is ¼ of the threshold. Cadmium, copper and zinc, usually assigned as metal pollutants from RAP do not pose any environmental concern in these aggregates [11]. Regarding soluble species sulphate is the main critical anion, particularly for CMC. Chromium and sulphate were also considered critical contaminants for concrete and mixed concrete recycled aggregates to be used in structural road layers by Galvín *et al.* [12]. Based only on the leachability criteria the low release of contaminants on the short term for the different recycled aggregates point to the feasibility of their use as alternative materials to natural aggregates in road construction. For RAP aggregates this conclusion opposes the given conclusion when total content of organic contaminants is considered.

Table 3: Contents of chemical species in eluates prepared according to EN 12457-4 [4]

Parameter [mg/kg, dry matter]	CMC	CRAP	MRAP	Limestone	Leaching limits
Cadmium, Cd	<0.005	<0.005	<0.005	<0.005	0.04
Chromium, Cr	0.126	0.046	<0.010	<0.010	0.5
Lead, Pb	<0.024	<0.024	<0.024	<0.024	0.5
Zinc, Zn	0.010	<0.006	<0.006	0.035	4
Copper, Cu	0.045	0.030	<0.014	<0.014	2
Nickel, Ni	<0.026	<0.026	<0.026	<0.026	0.4
Chloride, Cl^-	44	35	35	44	800
Sulphate, SO_4^{2-}	267	124	13	145	1000
Dissolved organic carbon, DOC	46	71	94	71	500
pH	11.29	10.95	9.67	7.90	–

The content of potential contaminants in the eluates prepared according to EN 1744-3[7} is shown in Table 4. The environmental risk of using EN 1744-3 [7] in a comparative context to EN 12457-4 [4] put in evidence the higher zinc, sulphate and chloride release in the former test. These differences were assigned to the influence of the pH and oxidation-reduction potential on controlling the solubility for different particle size distribution of the materials under test. RAP aggregates show lower release against crushed mixed concrete and a very similar release to the natural aggregate. This behaviour, underestimation of release, is in agreement with Van der Sloot and Mulder analysis regarding EN 1744-3 [8].

3.2 Lysimeter leaching tests

Table 5 lists the cumulative content of the analytes in the leachates from lysimeters. Lower and upper limits of release are reported for specific components whenever their concentrations in the leachate fractions were below the limit of detection.

It should be mentioned that milled RAP was combined with limestone aggregate to be in accordance with the composition used in the road pilot section constructed during the

SUPREMA project. The selected ratio, 30% MRAP/70% limestone, allowed fulfilling the grading envelope of the Portuguese Road Administration.

Table 4: Contents of chemical species in eluates prepared according to EN 1744-3 [7]

Parameter [mg/kg, dry matter]	CMC	CRAP	MRAP	Limestone
Cadmium, Cd	<0.005	<0.005	<0.005	<0.005
Chromium, Cr	0.103	0.022	<0.010	<0.010
Lead, Pb	<0.024	<0.024	<0.024	<0.024
Zinc, Zn	0.035	0.023	0.053	0.021
Copper, Cu	0.022	0.016	<0.014	<0.014
Nickel, Ni	<0.026	<0.026	<0.026	<0.026
Chloride, Cl^-	53	53	35	35
Sulphate, SO_4^{2-}	343	163	18	159
pH	9.96	8.12	8.14	6.86

Table 5: Cumulative contents of chemical species in leachates from lysimeters (L/S =5.0)

Parameter [mg/kg dry matter]	CMC	CRAP	MRAP/Limestone	Limestone
Cadmium, Cd	0 - 0.003	0 - 0.003	0 - 0.003	0 - 0.003
Chromium, Cr	0.083	0 - 0.025	0 - 0.025	0 - 0.025
Lead, Pb	0 - 0.005	0 - 0.005	0 - 0.005	0 - 0.005
Zinc, Zn	0 - 0.011	0 - 0.010	0 - 0.011	0 - 0.018
Copper, Cu	0 - 0.007	0.009	0 - 0.005	0 - 0.005
Nickel, Ni	0 - 0.016	0 - 0.015	0 - 0.015	0 - 0.016
Chloride, Cl^-	58	47	32	37
Sulphate, SO_4^{2-}	627	295	57	272
Dissolved organic carbon, DOC	21	37	14	7
Hydrocarbons (C10-C40)	–	0 - 0.15	0 - 0.15	–
pH	7.20-7.63	7.35-7.66	7.33-7.74	7.27-7.76

The most relevant factors regarding leaching from lysimeters are the pH, the redox conditions, the infiltration rate and volume, the exposed surface area and compaction. Table 5 shows the lower pH on the leachates from the lysimeters when compared to the eluates from the batch leaching tests which affect the release. In what concerns heavy metals, it is noteworthy that chromium is the metal showing higher content in the leachate from CMC, though lower for the batch leaching tests, which is compatible with lower leachate pH. It was also found a slight mobilization of copper in the lysimeter with CRAP, probably assigned to the high content of dissolved organic carbon of this aggregate. For MRAP similar effect was not seen probably due to the mixture of this aggregate with limestone

For the lysimeter with crushed mixed concrete sulphate is again critical showing higher content when compared to batch leaching tests. The leaching in lysimeters in the absence of the wearing layer corresponds to the worse situation whereby the increased release of soluble anions should not be regarded as the real behaviour in the field. According to Reid *et al.* [13], the wearing layer could reduce the water infiltration in 90%, therefore in undamaged pavements it is expected a significant decrease of sulphate leaching, corresponding to a lower environmental impact.

Towards clarifying the problems that could be related to the high total content of hydrocarbons C10-C40, an evaluation of these organic contaminants in the leachates from lysimeters containing RAP aggregates have shown that the release is always very low, below the detection limit (50 µg/l), corresponding to maximum leached content from 0 to 0.15 mg/kg. The released C10-C40 can represent up to 2% of the dissolved organic carbon which indicates that most part of the leached organic compounds did not come from the fraction C10-C40. In the literature similar low values of release of these hydrocarbons can be found for end-of-life tyres used as granulates in several civil engineering works [14]. Moreover, it is important to stress that the assessment of organic pollutants based on content is due to the lack of European leaching standards for these chemical species Therefore, it does not seem acceptable to restrict the use of RAP aggregates based on the fail of the criterion for the total hydrocarbon C10-C40 content when they pass all the remaining criteria for waste admissible in landfills for inert waste, as well as the established criteria in the Portuguese technical specifications for CDW to be used as recycled aggregates.

4. CONCLUSIONS

Based on the results from the leaching test, stated in the Portuguese Technical specifications, the release of contaminants from aggregates to be used in unbound pavement layers was higher for CDW aggregates when compared to natural aggregates. Nevertheless, the contaminants contents shows the feasibility of applying the different types of CDW since the chemical species analysed in the eluates comply with the limits for waste to be disposed in landfills for inert waste.

Regarding leachates from lysimeters, containing unbound recycled aggregates without a bituminous layer as wearing course, i.e., representing a totally damaged pavement owing to traffic and climate solicitations, the results for a ratio L/S = 5 show low release of contaminants. An estimation of the release for L/S=10 leads to contaminants contents below the admissibility criteria for waste into landfill for inert waste, in accordance to the laboratorial leaching test EN 12457 [4]. For intact pavements the contact with water occurs mainly by the shoulders of the road and a substantial decrease of leaching is foreseen, thereby strengthening the abovementioned viability of using the different recycled aggregates.

The critical constituents in the batch leaching tests were similar to those found in the leachates from lysimeters: chromium and sulphate for crushed mixed concrete aggregates and sulphate and dissolved organic carbon for reclaimed asphalt aggregates

In what concerns RAP aggregates, the high content of hydrocarbons C10-C40 had a minor release in leachates from lysimeters, which represents up to 2% of the dissolved organic carbon, and it should not hinder aggregate’s use. It is essential to establish leaching limits for organic pollutants to avoid restrictions based on the total contents since the released fraction maybe a very minor portion of the total content.

ACKNOWLEDGEMENTS

The authors acknowledge the financial support of FCT from Portuguese Ministry of Economy and Employment to the project “SUPREMA – Sustainable application of Construction and Demolition Recycled Materials (C&DRM) in Road Infrastructures”, and the Ambigroup SGPS SA and Teodoro Gomes Alho SA for materials supply.

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