Journal of Cleaner Production 66 (2014) 85–91

Contents lists available at ScienceDirect

Journal of Cleaner Production

journal homepage: www.elsevier.com/locate/jclepro

Leaching in concretes containing recycled ceramic aggregate from the sanitary ware industry

César Medina [a,*], Moisés Frías [b], María Isabel Sánchez de Rojas [b]

[a] Departamento Construcción, Unidad Asociada UEX-CSIC, Avenida de la Universidad, s/n, 10071 Cáceres, Spain
[b] Eduardo Torroja Institute for Construction Science (CSIC), C/Serrano Galvache, 4, 28033 Madrid, Spain

A R T I C L E I N F O

Article history:
Received 2 July 2013
Received in revised form
2 October 2013
Accepted 20 October 2013
Available online 5 November 2013

Keywords:
Leaching behaviour
Recycled concrete
Re-use
Ceramic waste
Water quality

A B S T R A C T

The re-use of construction and demolition, ceramic and similar waste in the construction industry has aroused considerable interest in recent years, as an avenue for furthering the sustainable use of resources and reducing the volume of waste dumped in landfills.

Recycling materials as components in the manufacture of cement-based products, however, calls for an understanding of the leachability of the elements present in the new materials that may be harmful to human health or the respective ecosystems. The present study addresses the effect of including recycled ceramic sanitary ware waste as a partial substitute (25%) for natural coarse aggregate in the manufacture of recycled concrete in direct contact with water intended for human consumption. The findings show that the inclusion of ceramic aggregate raises the alkali concentration (Na and K) slightly and lowers the concentration of other elements (B, Si, Cl and Mg) in the water. The levels of all the leached elements were observed to be lower than the limits specified in the legislation in effect on water for human consumption. Consequently, these new concretes are apt for use in such applications, for they ensure water quality.

1. Introduction

The need to set aside landfill space to accommodate the growing amounts of waste generated by the construction, ceramics and automobile industries is a serious environmental concern. Heightened public sensitivity to this problem has led to the implementation of new sustainability-based environmental policies that encourage the recycling and re-use of such waste (Delay et al., 2007).

The three keys to sustainability of construction materials are reduction, re-use and recycling (Marie and Quiasrawi, 2012).

The versatility of the cement and concrete industry affords it huge potential (Medina, 2011) to absorb new materials of varying origin as active additions in cement, as coarse or fine aggregates in mortar, concrete and road base/sub-base manufacture.

These recycled materials, such as construction and demolition (C&DW) or ceramic industry waste, are chemically inert (Galvin et al., 2012; Rodrigues et al., 2013). Nonetheless, depending on their origin, they may contain a certain proportion of hazardous elements, including metals (such as Znor Pb), anions (chlorides and sulfates) or organic compounds (polycyclic aromatic hydrocarbons) that may leach out when the materials are in contact with rain, surface or underground water. This constitutes a potential threat to the environment and for human health and safety (Dijkstra et al., 2012).

The requirements that must presently be met by all construction materials throughout their service life, in particular in connection with health, hygiene and the environment, are laid down in Directive 89/106/EEC on Construction Products (CPD) (Hjelmar et al., 2012). This directive will be repealed in 2013 by Construction Products Regulation no. 305/2011 (CPR), which calls for very meticulous assessment of recycled products containing substances that may have an adverse effect on human health.

The leaching resistance of granulated cement-based materials at the end of their useful life has been the object of considerable recent research (Delay et al., 2007; Meza et al., 2008; Engelsen et al., 2010, Galvin et al., 2012). The general consensus is that the leachability of substances from a given material is related to factors such as their solubility (Mulugeta et al., 2011). That, in turn, depends on other properties, including pH, the formation of inorganic complexes, the presence of dissolved organic matter and redox reactivity.

The intrinsic properties of materials that play a more or less prominent role in leaching can be divided into two categories: chemical factors such as pH or the chemical composition of the

* Corresponding author. Tel.: +34 927 25 72 03x51539; fax: +34 927 25 70 00.
E-mail address: cemedmart@yahoo.es (C. Medina).

0959-6526/$ – see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.jclepro.2013.10.029

aqueous phase in their pores, and physical factors such as permeability, release mechanism (diffusion), and form (monolithic or granular) (Van Der Sloot, 2000, 2002; Van Der Sloot and Dijkstra, 2004).

In recent years, Europe has made great strides in the harmonisation of its legislation on the assessment of substance leachability from monolithic or granular material to determine the applications for which materials can be used (Van Der Sloot et al., 2006). The background references for that endeavour included the existing European standards (NEN 7341, DEV S4, NFXP 31-211) and studies conducted by a number of researchers (Hohberg et al., 2000; Van Der Sloot, 2000; Sánchez de Rojas et al., 2004; Meza et al., 2008) to compare the variations in the findings when different methods are used.

As a result, European standard EN 12457 (CEN, 2002) is now the reference to be used to analyse leaching in granular waste and sludge, classified by particle size and liquid/solid ratio (L/S).Standard EN 14944 (CEN, 2007), in turn, is to be used to assess substance leaching from the factory-made cementitious products (concrete tanks and pipes) that carry and store water intended for direct human consumption, as well as the raw water from which drinking water is processed.

The present study constitutes a novel approach, given the paucity of international research (Marion et al., 2005; Hohberg et al., 2000) on the in-service leaching of substances from monolithic concrete and mortar. This question is of special relevance in new concretes that are in continuous contact with water intended for human consumption and that contain different types of recycled aggregates.

The use of waste from the ceramic sanitary ware industry as coarse aggregate to manufacture structural concrete has been widely studied by the authors of the present paper, who observed that concretes with 25% recycled aggregate have higher splitting tensile and compressive strength than the conventional material (Medina et al., 2012a,b, 2013).

The present study explores substance leachability in recycled concrete in which 25% of the natural coarse aggregate was replaced with recycled ceramic materials, based on the concentration of the leached material after various leaching periods. Research in this area is essential to determining the feasibility of applying these new concretes to uses that call for direct contact with water intended for human consumption, such as deposits or pipes.

2. Materials

Spain is the world's leading producer and exporter of ceramic sanitary ware, with a yearly output of seven million items (2008). The percentage of articles rejected for sale and thus discarded depends on the type of industry, as well as product requirements and other technical considerations. Rejects nonetheless account for 5–7% of total output according to data provided by manufacturers.

The recycled ceramic aggregate used to prepare the experimental concretes, supplied by a sanitary ware manufacturer, consisted of ceramic waste crushed with a jaw crusher to a size of 4/12.5 mm.

Table 1 lists the physical and mechanical properties of the coarse aggregates used in the present study. Both types conformed to the requirements laid down in European standard EN 12620 and Spanish code on structural concrete EHE 08 for the (natural or recycled) aggregates used in concrete manufacture.

The table shows that the ceramic aggregate had a higher pore volume and was consequently less dense than the gravel. With a pore volume similar to the value observed for ceramic electrical insulation, the recycled aggregate was 2.4 times more water-absorbent than the natural material. The flakiness index was eight times higher in the recycled ceramic aggregate than in gravel, primarily as a result of its outer morphology, in turn due to the original form of the waste and the crushing procedure. Another important property in aggregates is their Los Angeles coefficient. The recycled sanitary ware aggregate was found to be 39% more fragmentation-resistant than gravel (Medina et al., 2013).

Table 1
Physical and mechanical properties of the aggregates studied.

Property	Gravel	Ceramic aggregate	Specification for EN 12620
Real density of dry samples (kg/dm^3) – (EN 1097-3)	2.63	2.39	–
Water absorption (wt%) – (EN 1097-3)	0.23	0.55	<4.5
Flakiness index (wt%) – (EN 933-3)	3	23	≤35
Los Angeles coefficient (wt %) – (EN 1097-2)	33	20	≤40
Total porosity (vol. %) – (ASTM D 4404-84)	0.23	0.32	–

The chemical composition of the materials used to manufacture the concrete tested in the present study is given in Table 2. Note the substantial differences between the two types of coarse aggregate. The natural aggregate consisted primarily of silica (97.69%) with traces of other elements (Al, Fe, Ca, K and S), while the majority elements in the new recycled aggregate were silica (67.91%) and alumina (22.01%), with P, Pb, Cr and others as trace elements.

If these concretes are in contact with drinking water, the metals (Al, Fe, Mn, Cr and Pb), alkalis (Na) and halogens (Cl) present in the recycled ceramic aggregate might leach out, to the detriment of water quality (European Directive 98/83/EC and Spanish Royal Decree RD 140/2003).

Finally, the ceramic aggregate contained no organic material that would alter concrete setting or hardening rates nor did it exhibit alkali-aggregate reactivity, according to the petrographic study conducted.

3. Methods

3.1. Leaching test

The four-stage method described in European standard EN 14944-3 was used to assess substance leaching from cement-based products into the test water after exposure.

1) Four cubic specimens ($150 \times 150 \times 150$ mm^3) were prepared with the two types of concrete: reference concrete (RC) and recycled concrete containing 25% recycled ceramic aggregate

Table 2
Chemical composition of materials obtained by X-ray fluorescence.

Oxides (%wt.)	Cement	Natural aggregate	Ceramic aggregate
SiO_2	20.16	97.69	67.91
Al_2O_3	4.36	1.73	22.01
Fe_2O_3	2.52	0.13	1.41
MgO	2.21	–	0.29
CaO	63.41	0.04	2.41
Na_2O	0.35	–	1.91
K_2O	0.91	0.16	2.79
TiO_2	0.21	–	0.45
ZnO	–	–	0.16
Minority element (ppm)			
P	610.99	–	741.92
S	14297.71	8.88	280.35
Rb	0.01	–	45.72
Sr	–	–	84.56
Cr_{Total}	50.00	–	68.42
Pb	–	–	55.70
Cl	0.02	–	0.004
B	3.00	–	–
Mn	–	–	542.12