



Research article

Assessment of dynamic surface leaching of monolithic surface road materials

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ABSTRACT

Construction materials have to satisfy, among others, health and environment requirements. To check the environmental compatibility of road construction materials, release of hazardous substances into water must be assessed. Literature mostly describes the leaching behaviour of recycled aggregates for potential use in base or sub-base layers of roads. But little is known about the release of soluble substances by materials mixed with binders and compacted for intended use on road surface. In the present study, we thus performed a diffusion test with sequential renewal of water during a 64 day period according to CEN/TS 16637-2 specifications, on asphalt concretes and hydraulically bound monoliths, two common surface road materials. It is shown that release of dangerous substances is limited in these hydrodynamic conditions. It was particularly true for asphalt concrete leachates where no metallic trace element, sulphate, chloride or fluoride ion could be quantified. This is because of the low hydraulic conductivity and the low polarity of the petroleum hydrocarbon binder of these specimens. For hydraulically bound materials around 20,000 mg/m² of sulphate diffused from the monoliths. It is one order of magnitude higher than chloride diffusion and two orders of magnitude higher than fluoride release. No metallic trace element, except small quantities of copper in the last eluate could be quantified. No adverse effect is to be expected for human and environmental health from the leachates of these compacted surface road construction materials, because all the measured parameters were below EU (Council Directive 98/83/EC) or WHO guidelines for drinking water standards.

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1. Introduction

A number of studies have shown that runoff water collected in the vicinity of pavements have been polluted by many traffic-related sources (engine oils, pieces of tires, braking pads particles, guardrails, etcetera). Heavy metals (Pb, Zn, Fe, Cu, Cd, Ni) and hydrocarbons are some of the substances identified (Ball et al., 1998; Pagotto et al., 2000, 2001). But contribution of pollutants originating from road construction materials themselves to the runoff water quality has not been as investigated. For many years, a great emphasis was given to mechanical resistance and safety aspects of road construction materials. Since 2011, and the enactment of the new European Regulation for Construction Products, road construction materials also have to meet hygiene, health and

environment requirements. This issue is of particular concern for manufacturers of these materials. In relation with CE-marking, they indeed have to prepare a declaration of conformity for their products with the above-cited requirements prior to selling them within the European Economic Area. To check the environmental compliance of road construction materials, the release of hazardous substances from such products into water must be assessed. They may contain toxic substances that could be solubilized by percolating water and end up in nearby groundwater, and impact human and environmental health.

Most of leaching experiments have been performed on granular products to evaluate their suitability for usage in base or sub-base layers of roads. Natural rocks were tested (Tossavainen and Forssberg, 1999), but a greater emphasis has been placed in recycled aggregates (crushed concrete, reclaimed asphalt or industrial waste). Batch and column setups have been usually used. Results have shown that sulphate, chloride ions, Ba and Cr are the main substances released in leachates of recycled concrete aggregates (Arulrajah et al., 2013; Butera et al., 2014; Engelsens et al., 2009;

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Galvin et al., 2013; Kang et al., 2011). In leachates of reclaimed asphalt aggregates, As, Ba, Zn, fluorides have been quantified in the order of magnitude of 0.1–1 mg/kg of dry material (Legret et al., 2005; Somasundaram et al., 2015). PAHs between 1 and 5 µg/kg have also been detected (Brandt and De Groot, 2001; Legret et al., 2005). From these studies, most of the tested aggregates could be safely used in sub-base applications because concentrations of trace constituents rarely exceeded the regulatory values considered.

However, few studies have focused on the leaching behaviour of surface road materials (Kayhanian et al., 2009, 2010; Wahlström et al., 2000). They constitute the upper layer of pavement where aggregates are bound with bitumen, cement or lime. Literature has shown that the leaching of these monolithic products under normal conditions of exposure is essentially governed by diffusion or dissolution (Marion et al., 2005; Tiruta-Barna et al., 2004; Wahlström et al., 2000). Various leaching tests have been developed to study these mechanisms (Azizian et al., 2003; Coutand et al., 2011; Kayhanian et al., 2009; Wahlström et al., 2000). In the above studies, the hydrodynamic conditions (liquid to surface area-ratio, leachate renewal ...), as well as the duration of the experiments (from 24 h to several months) were rarely identical, making result comparison almost impossible. Therefore, the European Union through the CEN/TC 351 regulation has proposed a standardization of monolith testing with the Dynamic Surface Leaching Test (DSLIT), based on the NEN 7375 (2005). The DSLIT procedure is labelled TS 16637-2.

The present study is the first to assess the leaching of soluble substances from two surface road construction materials according to the specifications recorded in the TS version dated October 2013. We investigated both a cement based material and an asphalt concrete. The data provided in the paper may serve as a basis for identifying the amounts of substances that such road products may release in runoff water. This work also examined the dispersion of leach test results, because this point was not addressed in the TS 16637-2.

2. Material and methods

2.1. Sample preparation and characterization

Two road construction materials typically used as surface layer in France were examined: a hydraulically bound material and an asphalt concrete.

The hydraulically bound material was obtained by mixing 46% recycled concrete aggregates (0/6 mm) and 47% limestone (0/5 mm) with 7% hydraulic road binder enriched with activated blast furnace slags (Ligex FPL1, Calcia). The water soluble sulphate content was low in the recycled concrete aggregates (0.19% according to standard NF EN 1744-1). Six cylindrical specimens were manufactured using vibro-compression according to the standard NF EN 13286-52 in the Eurovia Company laboratories. The optimum water content and the maximum dry density were 10% and 2 t/m³ respectively. They cured 60 days in a closed vessel at constant temperature (20 ± 2 °C) and >90% RH before mechanical, permeability and leaching experiments were performed. Based on indirect traction resistance test values (NF EN 13286-42) and E-modulus (NF EN 13286-43), these road specimens were designed to tolerate between 50 and 150 heavy vehicles a day according to standard NF EN 14227-5.

Six cylindrical asphalt concrete specimens were prepared by mixing 94.7% aggregates (98% rhyolite and 2% limestone) with 5.3% 35/50 penetration grade bitumen, according to the standard NF EN 12697-12. They were prepared in the Colas group laboratories.

Three specimens of each material were used for the leaching

test and were characterized by their weight and geometrical dimensions (Table 1). The vertical permeability of the three other specimens was determined by measuring the infiltration rate of water through the specimens according to the standard NF EN 12697-19 (Table 1). The average permeability coefficient K of the specimens treated with a hydraulic binder was 3.68×10^{-6} m/s. They can be considered as moderately-permeable products. The average permeability of the asphalt concrete specimens was 2.12×10^{-7} m/s, similar to hydraulic conductivity of silty clay soils corresponding to low-permeable products.

2.2. Dynamic surface leaching test

Great care was given to the glassware cleaning (leaching vessels, filtration devices) to prevent any contamination from the environment.

For each leaching vessel, a blank test was first carried out. The vessels were filled with demineralized water. After 24 h, the leachate was renewed. Water was left in the vessels for another 24 h. Both eluates were collected and submitted for chemical analysis.

The dynamic leaching test was then performed in triplicate for each road materials, in order to assess the repeatability of the leaching procedure. Each test portion was placed on glass rods in a 12 L glass leaching vessel and the exposed surface A was completely submerged in demineralized water (Fig. 1). Water (resistivity of 15 MΩ cm) was introduced in the reactor such that the volume of liquid to exposed surface area ratio L/A of 8.0 ± 0.1 mL/cm² was met. It was kept in static conditions (Table 1). At least 20 mm space between the exposed surfaces of the test portion and the inner walls of the leaching vessel was respected. The reactor was closed to limit the CO₂ uptake from the air by the hydraulically bound material.

The leachate was renewed at predetermined time intervals (Table 2). At the end of each step, it was collected and analyzed. During the test, temperature was kept constant at 20 ± 5 °C.

2.3. Eluate analysis

The blank eluates and the 8 eluates collected during each leaching test were filtered over a 0.45 µm polyamide membrane. pH and conductivity were immediately measured (according to standards NF T90-008 and NF EN 27888 respectively). Concentrations of inorganic substances were determined by ICP-OES (NF EN 11885), atomic absorption spectrometry (NF EN 1483) and ion chromatography (NF EN ISO 10304-1): As (Quantification Limit QL = 0.01 mg/L), Ba (QL = 0.01 mg/L), Cd (QL = 0.002 mg/L), total Cr (QL = 0.01 mg/L), Cu (QL = 0.01 mg/L), Hg (QL = 0.0002 mg/L), Mo (QL = 0.01 mg/L), Ni (QL = 0.01 mg/L), Pb (QL = 0.01 mg/L), Sb (QL = 0.01 mg/L), Se (QL = 0.01 mg/L), Zn (QL = 0.01 mg/L), chloride (QL = 1 mg/L), fluoride (QL = 0.1 mg/L) and sulphate (QL = 1 mg/L). The concentration of Dissolved Organic Carbon (DOC) was also monitored (NF EN 1484) (QL = 0.5 mg C/L).

2.4. Expression of results

The eluate analysis provided the value of the concentration C_{ij} of each substance i per step j (in mg/L), with j varying from 1 to 8. The cumulative released concentration $C_{i,cum}$ for each element was then computed (in mg/L) (Eq. (1)).

$$C_{i,cum} = \sum_{j=1to8} C_{ij} \quad (1)$$

If the concentration of a substance in a particular eluate was

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