Materials and Structures (2023) 56:55
https://doi.org/10.1617/s11527-022-02078-5

ORIGINAL ARTICLE

Mechanical and leaching characterisation of impact-absorbing rubberised asphalts for urban pavements

Christina Makoundou · Alireza Fathollahi · Svein Kleiven · Stephen J Coupe · Cesare Sangiorgi

Received: 15 April 2022 / Accepted: 5 November 2022 / Published online: 20 March 2023

Abstract A new impact-absorbing material is being developed to protect vulnerable road users in urban areas and has been produced and tested, mechanically and environmentally in the laboratory. The main constituent of this innovative material is the rubber recycled from end-of-life tyres to foster a circular use of resources and exploit rubber's elastic properties. The study aims to provide a complete Uniaxial Compression Test (UCT) and leaching analysis of the material to propose and optimise a mix that is mechanically sound, durable, and respectful of the environment, in view of in-situ applications. Therefore, the UCT and Dynamic Surface Leaching Test (DSLT) were carried out on rubberised asphalt specimens with different mix designs. The 64 days cumulative concentrations of leached heavy metals and trace elements from unit surface of specimens were calculated and quantified, according to the CEN/TS 16637 standard. In parallel, thanks to a specific mechanical characterisation, compressive stress–strain curves were obtained, and the relaxation and elastic modulus were evaluated. The results from the

C. Makoundou · C. Sangiorgi (✉)
Department of Civil, Chemical, Environmental and Materials Engineering, University of Bologna, Via Terracini 28, 40131 Bologna, Italy
e-mail: cesare.sangiorgi4@unibo.it

C. Makoundou
e-mail: ngongo.makoundou2@unibo.it; christina.makoundou@ri.se

C. Makoundou
Department of Material and Surface Design, Research Institutes of Sweden (RISE), 114 28 Stockholm, Sweden

A. Fathollahi · S. JCoupe
Centre for Agroecology Water and Resilience (CAWR), Coventry University, Wolston Lane, Ryton-On-Dunsmore, Coventry CV8 3LG, UK
e-mail: alireza.fathollahi@strath.ac.uk

S. JCoupe
e-mail: aa0698@coventry.ac.uk

A. Fathollahi
Department of Civil & Environmental Engineering, University of Strathclyde, Glasgow, UK

S. Kleiven
Neuronic Engineering, KTH Royal Institute of Technology, Stockholm, Sweden
e-mail: sveink@kth.se

compression tests showed that the A-mixes have the best elastic and absorbing behaviour, especially those made with an SBS-modified bituminous emulsion (A4). The results from DSLT showed that the cumulative concentration of released elements, per unit surface of specimens were lower than the Dutch Soil Quality Decree (SQD) thresholds, taken as a reference. The low and early release of leachant observed for the mixtures, especially A4 as the most promising one, leave the possibility to handle the leaching with several solutions, including rubber coating treatment or water washing before their incorporation into the mix to limit and prevent their leaching while permitting very high injury reduction performances.

Keywords Crumb rubber · Rubberised asphalt mixtures · Cold and warm mixtures · Impact-absorbing pavement · Uniaxial compression tests · Dynamic surface leaching

1 Introduction

The injuries and trauma caused by falls occurring on pavements in urban areas are responsible for physical and psychological consequences of road users worldwide [1]. As the most exposed users, vulnerable road users (VRUs) are strongly impacted by this issue that is deemed to rise as the number of pedestrians and two-wheeled users increases [2, 3].

Nowadays, rubberised (recycled or not) surface layers and pavements are successfully used in playgrounds, sports fields and other civil engineering applications for their lower stiffness if compared to standard paving solutions [4–7]. In particular, to prevent injuries for the VRUs, protective surfacing, using innovatively and adapting advanced asphalt methods, to be used at a larger scale in urban and peri-urban areas (e.g. in the pavements of sidewalk and bike lanes) – thus without modifying already known processes and construction—are being developed to bring in impact-attenuation properties that are essential to reduce severe head, hips or shoulders injuries, including fractures, concussions, or internal bleeding [8] and their connected costs for the society at large. In line with this perspective, the proposed rubberised asphalt impact-absorbing material has been conceived and studied to meet this goal. Its full development will potentially enable a wide implementation in cities and wherever users' falls are more likely to occur while preserving the standard asphalt traditional production and construction methods. Moreover, using recycled rubber from end-of-life tyres as the main layer constituent fosters a circular economy approach instead of a linear one, allowing the recycling of high quantities of End-of-Life Tyre (ELTs) rubber [4, 9].

Preliminary studies have confirmed the feasibility and potential effectiveness of the impact-absorbing pavements (IAP) starting from various mix designs, using both warm [8, 10–12] and cold bituminous binders [13]; during their development process, a complete mechanical analysis of the material was required to assess their attenuation properties. The uniaxial compression test in different setups has usually been adopted to investigate the basic loading and relaxation behaviour of these materials [6, 7] because the methods commonly used for traditional asphalt, like the ITSM test (EN 12697–26), are not able to describe the considerable soft behaviour of the samples.

Furthermore, along with the material's mechanical performance, the environmental impact of the innovative paving solutions must be considered to design and build greener infrastructures [14, 15]. A recent study showed that the extraction, transportation and end of life stages of using virgin aggregates for pavement construction purposes have the largest contributions towards life cycle environmental and cost impacts [16]. This is the reason behind the objectives of many studies conducted worldwide aiming to replace virgin aggregates and binders with recycled materials, thus reducing the pressure on the environment. Crumb rubber from end-of-life tyres goes in this direction, and it is one of the most popular recycled materials to replace natural aggregates and improve bituminous materials characteristics in pavements.

Up to now, limited scientific studies have investigated the behaviour of crumb rubber in contact with water and its potential to release heavy metals and trace elements [17], or reported the concentrations of leached As, Ag, Ba, Cd, Cr, Hg, Pb, and Se from tyre rubber used in artificial turfs [18], or examined the zinc leaching from tyre crumb rubber through column tests for a duration of 96 h. The latter study reported an initial pulse of elevated zinc leaching of 3 mg/L before settling down to a steady-state value of 0.2 mg/L. Another study on crumb rubber samples from artificial turf playgrounds showed that the granulates contained up to 500 mg/kg of aluminium [19]; while concentrations of cobalt, cadmium, copper, sodium, magnesium and iron in crumb rubber granulates from artificial playgrounds have been reported by other literature [20–24]. Nonetheless, the systematic assessment of the release of heavy metals and trace elements from asphalt specimens containing crumb rubber granulates in contact with urban runoff is not common. For this reason, the present study aims to evaluate the leaching performance of the rubberised asphalt – and is this case highly rubberized impact-absorbing asphalt – according to the Regulations recognised by the European Commission (EC). In fact, more than one standard procedure has been identified by the EC to investigate the leaching behaviour of construction materials, including the CEN/TS 16637–2:2014 or the CEN/TS 14405. In particular, the Dynamic Surface Leaching Test (DSLT) CEN/TS 16637–2:2014 standard aims to determine the release of "Regulated Dangerous Substances" from construction products into soil, surface water and groundwater and it has been utilised by several studies to evaluate the leaching behaviour of construction materials [25–30]. The results from the leaching tests should be compared with the thresholds set by the EC or other regulatory bodies including the Water Framework Directive (WFD) [31] to ensure the safety of materials under investigation to be used in the urban environment.

In the light of the above, this paper wants to provide a complete uniaxial compression and leaching data analysis from the proposed innovative impact-absorbing material, thus contributing to the developing research steps of a promising paving solution.

Six mixes were designed using two different percentages of crumb rubber granulates and four different binder types. Axial relaxation, recovery abilities and compressive strength values were obtained and compared to known playgrounds or rubber composites with sufficient shock-absorbing performances. In the leaching experiments section of the present study, the release of heavy metals and trace elements from the six rubberised asphalt concretes were evaluated according to the mentioned CEN/TS 16637–2:2014 standard.

2 Methodology

2.1 Experimental plan

An overview of the study plan, including the description of different materials and resources used, the laboratory preparations and productions of samples, and the corresponding characterisations carried out, namely the DSLT and the UCT, is shown in Figure 1. Further details are explained in the following section.

2.1.1 Rubberised impact-absorbing asphalt mixture

Six types of rubberised IAP mixtures were produced. The dry process consisting in mixing the aggregates and binder before adding the crumb rubber was used. All the mixtures contain, as the main constituent, the crumb rubber from ELTs. The rubber was obtained from a company treating waste tyres using the common ambient shredding process. In all the proposed mixtures, the size distribution of rubber is 0–4 mm and 2–4 mm, as shown in Table 1.

Two main mixes containing different recycled rubber amounts were assessed: the A-mixes with 56% rubber (by volume of the total mix) and the B-mixes with less rubber (50%). The natural aggregates mix constituent were 0–14 mm limestone aggregates, which were partially replaced by the rubber particles (in volume). In the A-mixes, the aggregates are partially substituted by the same size of rubber granulates, while in the B-mixes, a lower percentage of the largest size aggregates (10–14 mm) is adopted. Finally, four different bituminous binders were used: two commercial asphalt binders, including a Styrene–Butadiene–Styrene (SBS) modified bitumen purposely designed to incorporate rubber using the dry method (1), and a standard 50/70 penetration grade neat bitumen (2); and two cold bitumen-based emulsions (with the same residual binder content), a Latex modified one (3), and an SBS modified one (4). The SBS content is a proprietary information of the producing company. Usually, it ranges between 3 and 7% depending on the level of modification. In this case higher values are used as the binder is considered to be a "hardly-modified" bitumen.

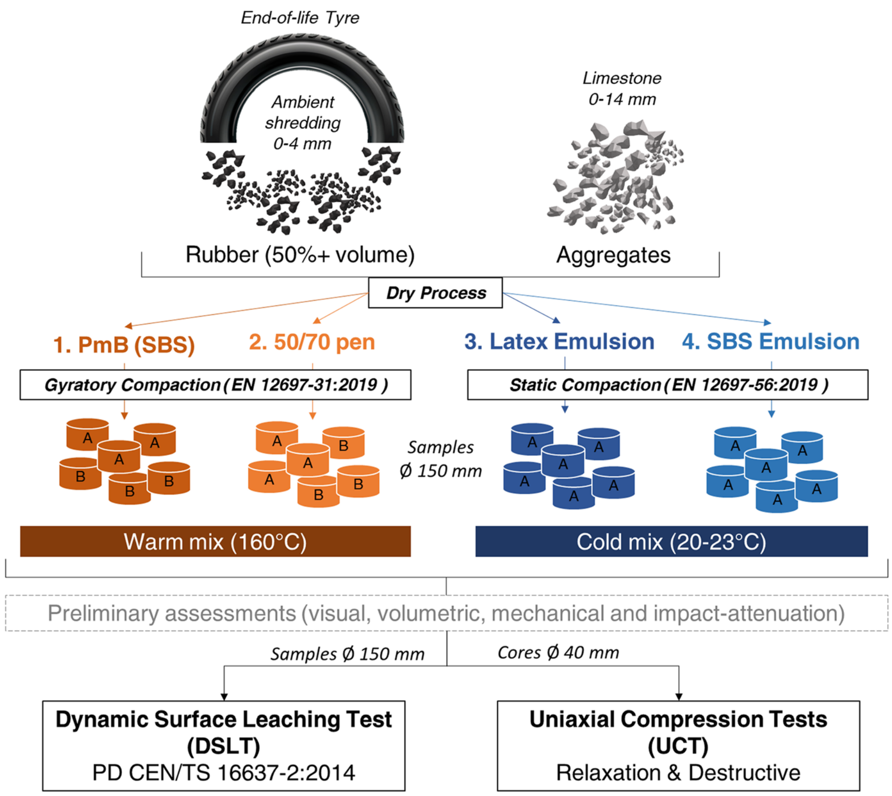


Fig. 1 Experimental plan of the study. The scheme describes the raw materials used, including the rubber, the aggregates, and the different binders. The dry process was used to produce different specimens compacted by means of gyratory and static compaction. These were tested with the DSLT and UCT tests

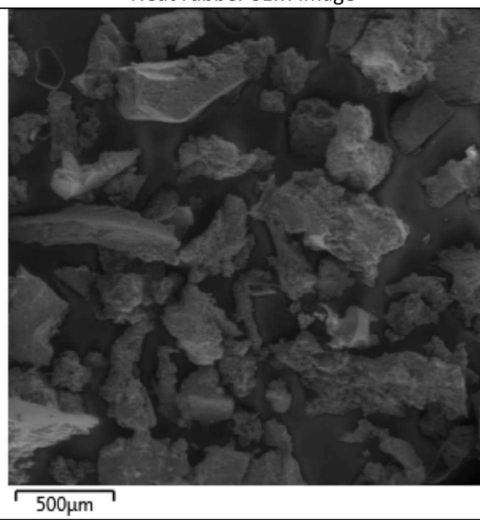
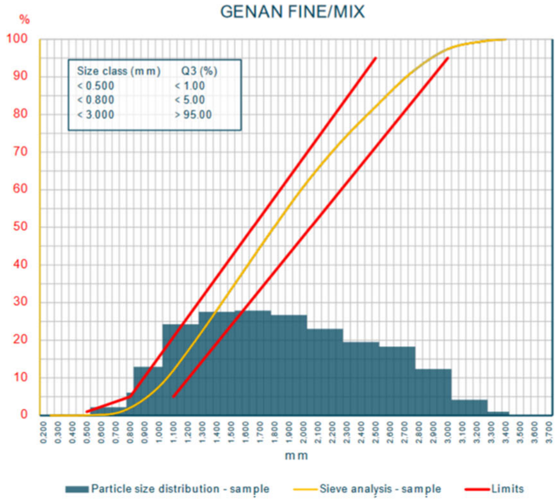
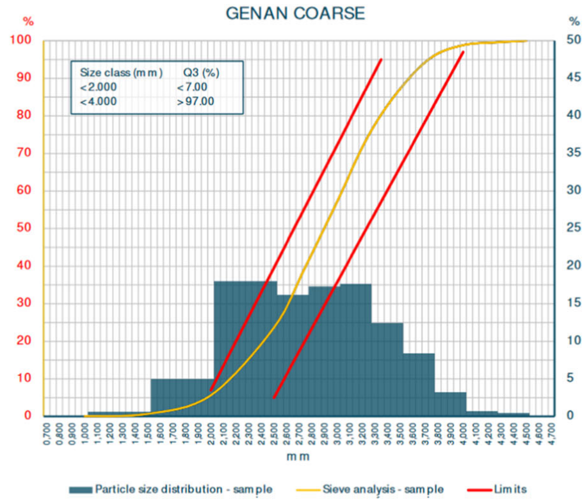
2.1.2 Production of samples

The bituminous samples of 150 mm in diameter and 40 mm in thickness were produced following two main compaction methods: the gyratory compaction (EN 12697–31:2019) and the static compaction (EN 12697–56:2019), adopting the last one for cold bituminous mixes. The mixing times were defined using the organoleptic assessment of mixtures (EN-12697–55:2019) to produce well coated and homogenous mixtures. The hot/warm mixes were gyratory compacted with 80 cycles at approximately 80 °C (A1 and B1) or 50 °C (A2 and B2) permitting a correct adhesion of all the components. Instead, the cold materials were statically compacted at approximately 20 °C (room temperature). These cold samples were subjected to curing after being extracted from the mould and cured at room temperature. The leaching tests were conducted on the full original samples, while the stress-relaxation and compression tests required coring the samples to a smaller diameter to allow testing. Therefore, the original samples were cored using a 40 mm coring rig, as shown in *Fig. 2*.

2.1.3 Testing criteria

This study is part of a research project aimed to develop a new material for users' protection purposes, and that allows recycling end-of-life tyres in large quantities. A preliminary laboratory study was recently conducted by the authors in which a set of initial physical and mechanical characteristics were determined on the impact-absorbing materials. In parallel, a Head Injury Criterion (HIC) testing investigation [33, 34] was conducted on the same materials. All recorded data are listed in Table 2 and the same nomenclature has been used for the mixtures on which stress-relaxation and compression tests have been performed to allow a more specific mechanical analysis that could also be used for Finite element

Table 1 Neat rubber Scanning Electron Microscope (SEM) image and granulometry values from [32]. The table contains a SEM picture of the rubber mix and the granulometric data of the two adopted rubbers, allowing a rubber size distribution of 0–4 mm [32].

Neat rubber SEM image	
	
Percent Retained per sieve [%]	
"Fine mix"	**"Coarse"**
	

method (FEM) modelling purposes. In addition, the chemical analysis outcomes, and their potential influence on the environment (in specific on water) are very important for the comprehensive assessment and validation of the proposed material.

This paper aims to investigate the mechanical uniaxial-compressive characteristics and analyse the leachates in water, estimating the potential environmental impact of the six rubberised mixtures under assessment. These data will be used to optimise the material constituents to maximise the impact-absorbing properties, maximise the amount of recycled rubber, and minimise the contamination of waters, while keeping the production and laying processes feasible at an industrial scale. The latter will have to be assessed with an ad hoc experimental site investigation already foreseen for the material development.

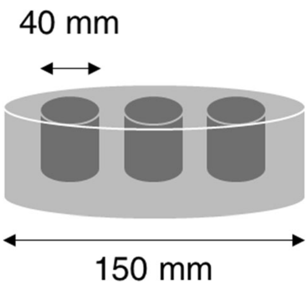


Fig. 2 Scheme illustrating the 40 mm cores obtained from 150 mm samples for the uniaxial tests

3 Characterization

3.1 Mechanical test methods and analysis

The study focuses on the compression of samples. In the case of compression, the obtained results can be associated to the actual axial load on the material and to the foreseen injury reduction modelling and simulation. The choice of the authors was to limit this study to the UCT. However, also direct tensile and torsional tests have been conducted however the poor adhesion of the highly granulated surface of the materials, to the testing machine was giving uncertain results; while the shear tests are foreseen, as it can also characterize the behaviour of the newly developed material. Therefore, UCT (relaxation and destructive) were performed at room temperature (20 °C) on the 40 mm diameter cylindrical specimens using an Instron Electropuls E3000 testing machine (Fig. 3) at the KTH Neuronic Laboratory, Sweden. Trial tests have been performed to optimise the initial deformation allowing to record the complete relaxation of samples and the required displacements to reach failure, without overcoming the compressive load limit of 3kN.

Various material parameters such as the elastic modulus, the elastic recovery and the mechanical behaviour are determined and shown as stress–strain curves illustrating the hysteresis during the testing phase Fig. 3.

3.2 Uniaxial unconfined stress-relaxation test and calculations

After the initial calibration phase, some relaxation tests were carried out. Samples were compressed at approximately 5% (2.5 mm) strain with a load application speed of 20 mm/s. When reaching the target strain, the load was held for 60 s, after which the unloading phase was carried out at 0.6 mm/s. Additionally, an elastic recovery test was carried out to verify the applied strain and assess the sample's relaxation in time from 1 to 30 min after the release of the initially applied stress. The machine-recorded parameters allow the calculation of the Relaxing Stress and Strain (Engineering), and of the Relaxation Modulus.

$$\text{Relaxing Stress [MPa]} = \frac{\text{Load } [N]}{\text{Original cross sectional area } [\text{mm}^2]} \tag{1}$$

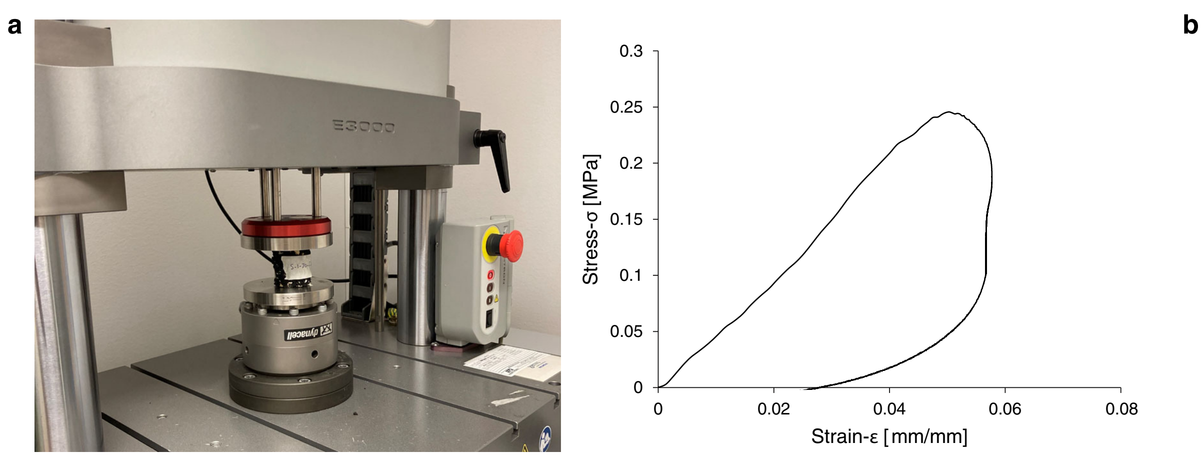


Fig. 3 **a** Compressive tests setup; **b** Stress–strain graph of a stress-relaxation test illustrating the hysteresis during the relaxation phase

$$\text{Strain} = \frac{\text{Displacement [mm]}}{\text{Sample Initial Height [mm]}} \quad (2)$$

$$\text{Relaxation Modulus [MPa]} = \frac{\text{Relaxing Stress [MPa]}}{\text{Strain}} \quad (3)$$

Then the Relaxation Modulus G(t) for a linear viscoelastic model (Standard linear solid) was fitted through a MATLAB script using the following equation:

$$G(t) = G_\infty + (G_0 - G_\infty)e^{-\beta t} \quad (4)$$

where: $G(t)$ = Relaxation Modulus Curve Approximation using 1 term. G_0 = Initial Relaxation Modulus. G_∞ = Final Relaxation Modulus. β = Time relaxation constant (s^{-1}).

3.3 Axial unconfined destructive compression procedure

The compression strength test was conducted at two different application speeds, corresponding to different strain rates, both quasi-static and dynamic. To calculate the speed of application, starting from the strain rate, the following relation has been considered:

$$\dot{\varepsilon}[1/s] = \frac{v[mm/s]}{h[mm]} \Rightarrow v = \dot{\varepsilon} \times h \quad (5)$$

with the strain rate $\dot{\varepsilon}$ being equal to:

- 0.01 [1/s] (v = 0.01.AVG H_0 = 0.43 mm/s) – Quasi-Static (QS)
- 1 [1/s] (v = 1.AVG H_0 = 43 mm/s) – Dynamic (*D*)

The destructive uniaxial compressive test was carried out and samples were compressed at approximately 50% (25 mm) strain with an application speed of 0.43 (QS) and 43 (*D*) mm/s. When the maximum displacement of 25 mm was reached, the load was not held and the unloading phase was carried out at a speed of 0.43 (QS) and 43 (*D*) mm/s. Compressive Engineering stress, Engineering strain and Elastic modulus are calculated as follows.

$$\text{Compressive Stress [MPa]} = \frac{\text{Load } [N]}{\text{Sample area [mm}^2]} \quad (6)$$

$$\text{Strain} = \frac{\text{Displacement [mm]}}{\text{Sample Initial Height [mm]}} \quad (7)$$

$$\text{Elastic Modulus [MPa]} = \frac{\text{Compressive Stress [MPa]}}{\text{Strain}} \quad (8)$$

3.4 Dynamic surface leaching test

The leaching behaviour of rubberised asphalt specimens was evaluated based on the CEN/TS 16637–2 standard for construction and building materials. Rubberised samples for DSLT were of 150 mm in diameter and 40 mm in thickness and compacted according to EN 12697–31:2019 and EN 12697–56:2019 standards. The leachant in DSLT was deionised water with a conductivity less than 0.5 mS/m according to EN ISO 3696. A 0.1 mol/L nitric acid was used for rinsing purposes (Fig. 4).

A set of glass vessels with lids were used as leaching tanks to avoid prolonged contact with air and possible CO_2 uptake. Rubberised asphalt samples were placed in glass vessels with a minimum of 20 mm distance between specimens and glass vessels in all directions. Small glass pieces were placed under the specimens to avoid direct contact of asphalt surfaces and leaching vessels and provide a direct contact between the bottom side of specimens and leachant. A representative leachant sampling procedure was performed according to CEN/TS 16637–1 standard. The temperature of the lab was kept constant

Fig. 4 Glass vessel containing the rubberised asphalt in direct contact with the water

between 20 and 25 °C. A L/A ratio of 80 was selected for the leaching tests and the volume of leachant was calculated according to the surface of specimens. The DSLT leaching tests were carried out in triplicates for all rubberised asphalt specimens mix designs. Three glass vessels filled with deionised water and no asphalt specimens were assigned as controls to detect and eliminate the contamination from lab equipment and the surrounding environment. Samples from control replicates had to have less than 0.2 mS/m and elemental concentrations below the detection limit to fulfil the minimum requirements, otherwise, DSLT experiments had to be repeated. Leachants from control and asphalt specimens were sampled and renewed after 0.25, 1, 2.25, 4, 9, 16, 36, and 64 days from the experimental setup. Therefore, the leaching tests comprised of 8 stages with durations of 0.25, 0.75, 1.25, 1.75, 5, 7, 20 and 28 days.

Electric Conductivity and pH of the leachants were measured at the end of all stages of leaching tests according to the EN 16192:2011 standard, using an EC and pH meter. Leachant samples were then filtered off-line using 0.45 µm membrane filters. Samples were then centrifuged at 2000 g for 5 h before storage at 5 °C and elemental analysis. The concentration of heavy metals and trace elements in filtered samples from DSLT was evaluated using a Perkin-Elmer optima 5300 DV Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES) instrument. ICP-OES is an analytical chemistry technique used to determine concentrations of specific elements in samples. The elements under investigation in the present study included Al, As, B, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Mo, Na, Ni, Pb, Sb, Se, Si, Tl, V and Zn. Analytical methods for quantification of mentioned elements were developed and commissioned for the ICP-OES instrument. After quantification of concentrations of described elements in leachants from different stages of DSLT, cumulative leached concentration for each element was calculated according to CEN/TS-16637-2 2014 as follows:

$$R_n = \sum_{i=0}^{n} r_i \tag{9}$$

where R_n (mg/m^2) is the cumulative area release of the substance for period n including fraction i = 1 to n of DSLT test, r_i is the area release of the substance in fraction i, and i is the DSLT stage. SPSS 21.0 software (Chicago, Illinois, US) [35] was used to perform statistical analysis of data from DSLT.

4 Results and discussions

4.1 Relaxation of the samples

4.1.1 Elastic recovery of the materials

The materials' recoverability after applying a stress was tested and recorded at different time. Figure 5

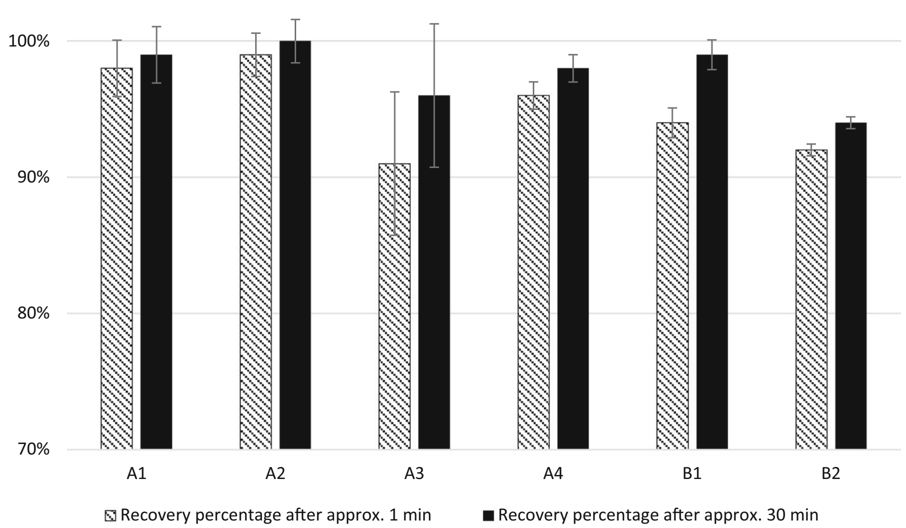


Fig. 5 : Average elastic recovery values for the maximum applied strain

Table 2 Impact-Absorbing Pavements made with cold and hot asphalt binders and recycled rubber: summary of the preliminary characteristics of the mixtures

Mixture	A1	A2	A3	A4	B1	B2
Rubber volume (Total mix) [%]	56	56	56	56	50	50
Bitumen type	PmB-SBS	50/70 pen	E-Lat	E-SBS	PmB-SBS	50/70 pen
Bitumen volume (Total mix) [%]	15	15	15	15	15	15
Bulk Density *EN 12697–6*	1.351	1.292	1.441	1.364	1.533	1.256
Voids content [%] *EN 12697–8*	4	3	5	8	3	6
Indirect Tensile Stiffness Modulus [MPa] (5 °C) *EN 12697–26*	230	266	148	144	526	209
CFH at HIC* 1000 [m] *EN 1177*	0.93	1.00	1.22	1.66	0.74	1.08
Natural ageing resistance **	High (7/8)	Weak (3/8)	Weak (4/8)	High (7/8)	High (8/8)	Weak (8/8)
Externalities** (fumes and smells, workability)	High (2/8)	High (3/8)	Low (7/8)	Low (8/8)	High (2/8)	High (3/8)

*Through the measurement of the severity of a head injury that is likely to arise from an impact, i.e. Head Injury Criterion (HIC), using an artificial concentric head (approx., 5 kg in mass simulating a simplified version of adult human head) containing accelerometers and sensors, it is possible to calculate the Critical Fall Height (CFH) of the pavement that is the maximum height from which a fall on the paved surface will have a sufficient level of impact attenuation

**To evaluate the durability and the effects of natural weather, the analysis was performed before and after a Natural Ageing Procedure (NAP) of 3 months under light−exposed, cold, and humid weather conditions. Temperatures ranged between 15 and −7 °C, with 82% of humidity on average, including several rain and snow events and an estimated Ultra−Violet (UV) index of 2–3 corresponding to a low to moderate exposure level were reported. Using organoleptic, volumetric and mechanical testing and mark (from 1 to 4) have been attributed to the mixtures assessing the: fume and smell; the workability of the mix, the cohesion of the samples, the durability of the mix after the NAP and the injury prevention performances (assessment marks: 1: very weak; 2: weak; 3: high; 4: very high.). In the case of the natural ageing resistance, the cohesive properties of the samples and the durability after the ageing were used (8 point in total). The samples that were fully degraded and unexploitable after 3 months of ageing (falling apart, malleable) were considered weak (A2, B2 and A3), while A1, B1 and A4, resistant. The lower is the mark, the less resistant is the material. In the case of the externalities, the fume and smell and workability categories are assessed on 8 points. The lower is the mark, the higher are the impacts related to the externalities

demonstrates the recovering abilities of the different mixtures after a compression applied to deform of approximately 2.5 mm.

When using the same binder but different rubber amounts (A1-B1 and A2-B2), the more rubber is contained in the mix, the faster are the material's recovery abilities. After 1 min, A1 recovers at 98% while B1 at 94%, but after 30 min, they reach 99% of the recovery. In the case of binder 2, this difference is even more visible as, after 30 min, B2 is still not recovered at 95%.

The B2 mix show a very low recovery capacity, and this represents an issue concerning future applicability in real conditions considering that daily stress will be applied to the material.

For the A-mixes, binder 2 show better recovery values however, concerning the B-mixes, binder 1 display better recovery values. Both the rubber and the binder affect the elastic recovery of the samples.

At an equal rubber amount (A1, A2, A3 and A4), the hot-made samples recover faster and better than the cold-made samples. This can be explained by the samples' structure that has less air voids than A3 and A4 (Table 2). Additionally, for the same reason, the compaction method can impact this result (Fig. 1).

A3 shows the lowest recovery possibilities. In fact, after 1 min of compressive load applied, the samples A1, A2 and A4 are already recovered at more than 95% of the initial height while for A3, the 95% recovery is obtained just after 30 min. A3, like B2, doesn't have enough recovery potential and can lead to a fast deterioration of the material if applied on a sidewalk/bike lane portion. These were also observed in preliminary ageing results as reported in Table 2, in

Table 3 Summary of the G_0, G_{inf} and β value and standard deviation of G for each mixture

Mixture	G_o	G_{inf}	Average of standard deviation of the elastic modulus	β
A1	4,659	2.666	0.7649	0.1472
A2	2.4066	1.1035	0.5666	0.1220
A3	1.4427	0.2897	0.2696	0.1166
A4	2.435	0.8809	0.3655	0.1296
B1	15.3679	4.8464	1.3237	0.4776
B2	1.8782	0.6822	0.2927	0.1300

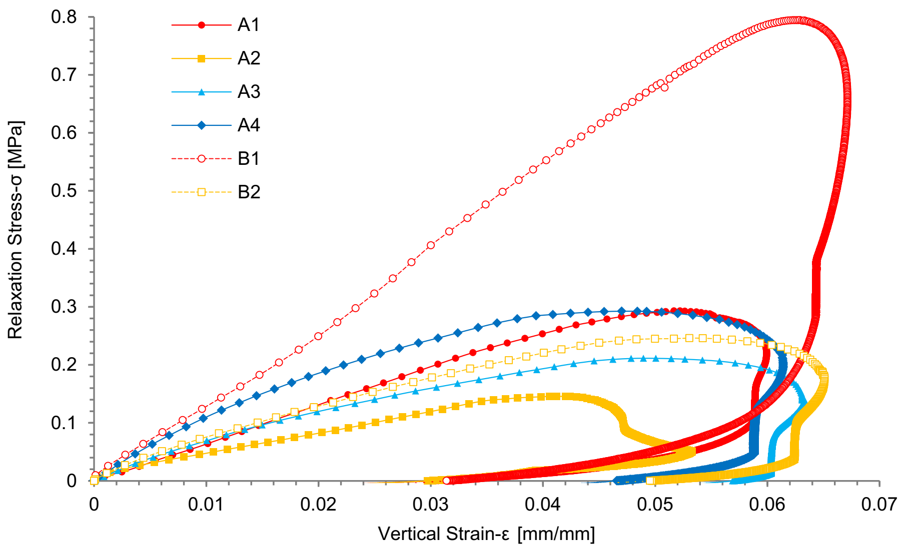


Fig. 6 Relaxation Stress-σ and Vertical Strain-ε curve

which A2, B2 and A3 were the least resistant materials and A1, B1 and A4 the more resistant ones. This observation is not verified by A2 elastic recovery. Additionally, the elastic recovery possibilities seem not to affect the HIC results because A3 has a high CFH and a poor recovery performance, A1 has a low CFH, but good recovery and A2 has a high CFH and a high recovery. As a matter of fact, it is difficult, through this test, to anticipate the contribution of the recovery on the injury reduction measured with an HICmeter.

4.1.2 Stress–strain analysis and calculation of the relaxation modulus

MATLAB was used to calculate the relaxation moduli of the six asphalt mixtures according to Eq. (4). Figure 6 shows the test results used for the relaxation moduli calculation for the six asphalt mixtures.

The stress–strain curves have a similar shape apart from the A2 mixture one. B1 and B2 have larger hysteretic areas than A1 and A2, respectively; the lower rubber content can explain this in the B-mixes. The hysteretic loss is similar for A1 and B1 but very different for A2 and B2, with A2 recording more hysteretic loss. As for the A-mixes, the hysteretic areas from the higher to the lower can be listed as follows: A4 > A1 > A3 > A2. A1 and A2 record the lower hysteretic loss and A3 the higher. This confirms the elastic recovery observed in Fig. 5.

B1 relaxation stress is much higher than all the other mixes. Figure 5 also described a high possible deformation but with and high capability to recover.

In general, the mixture made with SBS modified binder (A1, A4 and B1) allows the specimen to have a noticeable deformation and maximum recovery after compression. However, this is not observable with the mixture made with the other binders.

Additionally, the non-modified binder and emulsion (numbers 2 and 3) lead to lower moduli. At the same rubber amount, mixture A1 provides the highest modulus and A3 the lowest. The type of binder used

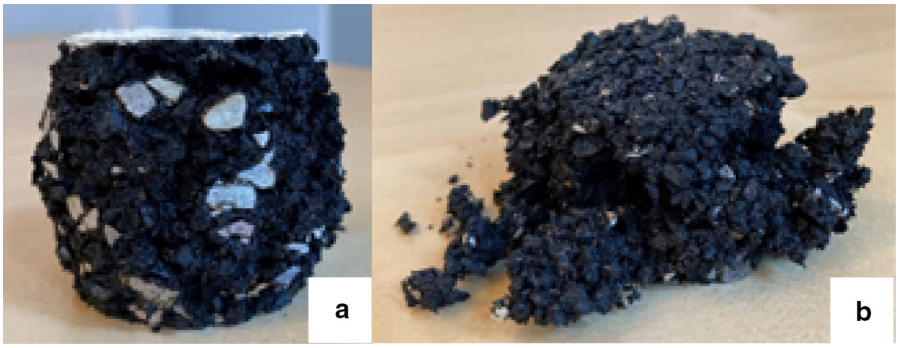


Fig. 7 Typical failures after destructive testing. **a** sample fails with no loss of cohesion (A1, A4 and B1); **b** sample fails with loss of cohesion (A2, A3 and B2)

has a higher influence on the material behaviour than the rubber amount. In fact, B2, even if containing less rubber, is more deformable than A1 or A4 and it is less likely to spring back to its original shape faster after loading. This data validates the results observed in previous investigations including the outcomes of the elastic recovery. B2, A3 and A2 are less likely to resist daily stress than A1, A4 and B1.

4.2 Failure behaviour and compressive strength

4.2.1 Failure analysis

Two main sample failure behaviours have been registered during the compression strength study. These are shown in Fig. 7 During the destructive UCT process, it was found that samples easily fail along the interface between the rubber-binder matrix and mineral aggregate, no matter the binder type.

The samples made with the SBS-modified binders had still links after the compression (Fig. 7, Fig. 7a) while the samples made with the other binders scrambled without valuable link in between the different components (Fig. 7, Fig. 7b). These observations confirmed the preliminary visual assessment on the cohesive aspect of the samples and the results of the relaxation tests. It was already seen that the samples made with binders 2 and 3 had the worst resistance to weather conditions and they are less likely to be used as impact-absorbing pavements in real conditions, due to their weak durability in time.

Also, this analysis confirmed the recovery and relaxation test results in which A3, B2 and A2 mixtures were less likely to be selected as valid IAP materials than A1, B1 and A4 mixtures.

4.2.2 Stress–strain curve analysis and calculation of the elastic modulus

The Elastic moduli of the six asphalt mixtures were calculated with MATLAB and according to Eq. (8) for the quasi-static and dynamic test. Values are shown in Fig. 8.

According to Fig. 8 the stress–strain behaviour of the samples is similar to the one observed on playgrounds [6, 36]. The stress similarly ranges from 0 to 3 MPa. B1, A1, A2 and B2 samples could be reproducing a usual playground behaviour, while samples A4 and A3 are considered softer, and by extension, safer [6, 36]. Figure 8

For various rubber amounts, the 1-mixes and 2-mixes have totally different stress–strain curves trends, but A1 and B1 curves have similar behaviour corresponding to harder materials and A2 and B2 corresponding to softer materials. These values confirm the CHF values collected with the HICmeter and described in Table 2. B1 registers higher stress than A1, however A2 has a higher stress value than B2. This can be explained by the crack and destruction configuration in which the asphalt composite is left without bonds and rapidly, the calculated stress is attributed to aggregates interaction instead of the full material.

The strength rank as A2 > A1 > A4 > A3 for the same amount of rubber. The A1 and A2 curves are different, while A3 and A4 are similar. A3 and A4 curves corresponds to very soft materials. It also

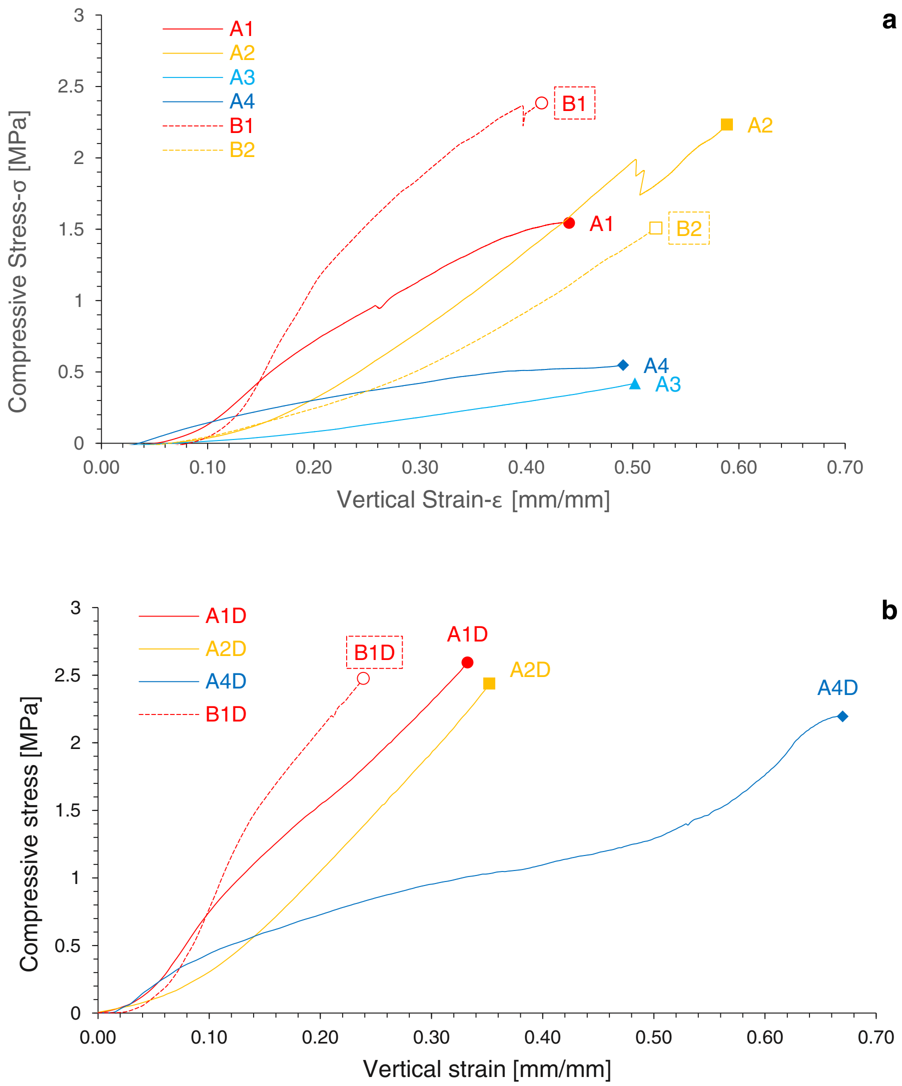


Fig. 8 Quasi-static (**a**) and Dynamic (**b**) Compressive Stress-σ and Vertical Strain-ε curves

confirms the CFH values as A3 and A4 have the higher CFH, thus seems safer against injury reduction, especially fractures caused by the hard surfaces.

The dynamic study of the failure behaviour of the sample gives a more evident result regarding the characteristics of the mixtures. Due to a lack of cohesion of the samples during the coring, the A3 and B2 samples could not be assessed. This observation also confirms the results discussed in the previous sections This limits the study to A1, A2, A4 and B1 mixtures, which can be considered good alternatives to be applied in an in-situ trial.

At equal binder but different rubber amounts, the difference in the stress to failure is low. A4 has a higher rubbery behaviour than A2 and A1 and its deformation is higher at equivalent stress. This shows better energy-absorbing behaviour toward stress.

Thanks to these results, A4 seems the best compromise between the resistance and the softness with regards to injury reduction. In fact, B1 and A1 are considered too hard and stiff, while A2 B2 and A3 not resistant and do not recover enough for such an application.

4.3 Dynamic surface leaching test

4.3.1 Electric conductivity and pH

The pH and electric conductivity of leachants in contact with different rubberised asphalt mix designs during

DSLT experiments were measured on day 0.25 to 64 according to the CEN/TS 16,637–2 standard and are presented in Fig. 9. The highest mean electric conductivities for mix design A1, A2, A3, A4, B1 and B2 were 986 (Day 16), 1071 (Day 9), 920 (Day 16), 962 (Day 4), 823 (Day 64) and 826 (Day 36) (uS/cm), respectively. As results suggest, B-mix designs showed lower electric Conductivity than A-mix designs. Moreover, the maximum electric Conductivity occurred at earlier stages of DSLT (Day 4–16) for A-mix designs compared to those of B-mix design (Day 36–64). This observation revealed that A-mix designs with a higher content of crumb rubber particulates released higher total ions in shorter time into the leachants during the DSLT. The electric Conductivity of control replicates was lower than 0.2 mS/m, which satisfied the first quality check of the DSLT experiments.

The pH evolution of leachants from DSLT experiment 6 types of rubberised asphalt are presented in Fig. 9b. According to the results, the pH of leachants from all specimens showed a constant increase as the duration of contact increased. On days 0.25 and 1, the pH for all specimens stayed below 7.5 pH of all specimens increased from less than 7.5 to above 8 between day 1 and 36. However, the maximum pH (> 8.5) was observed at the last stage of DSLT experiment were

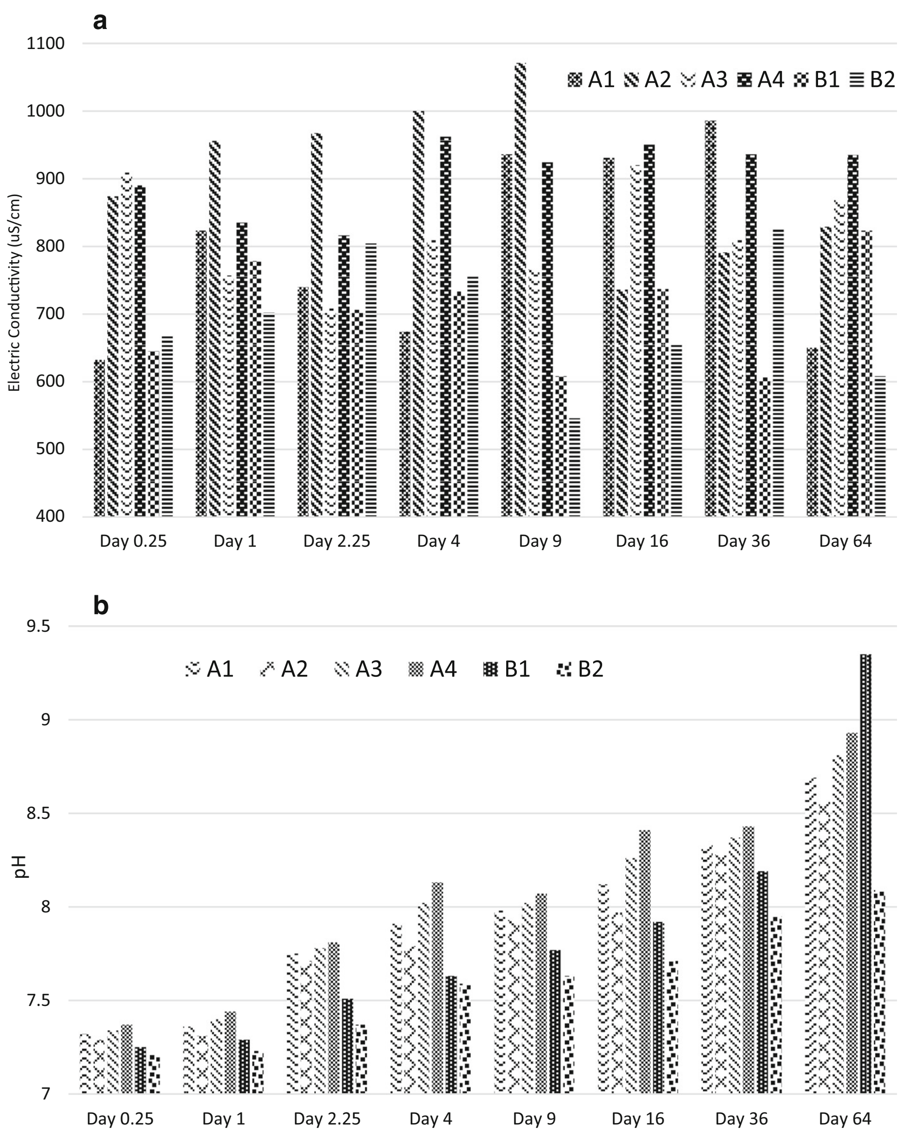
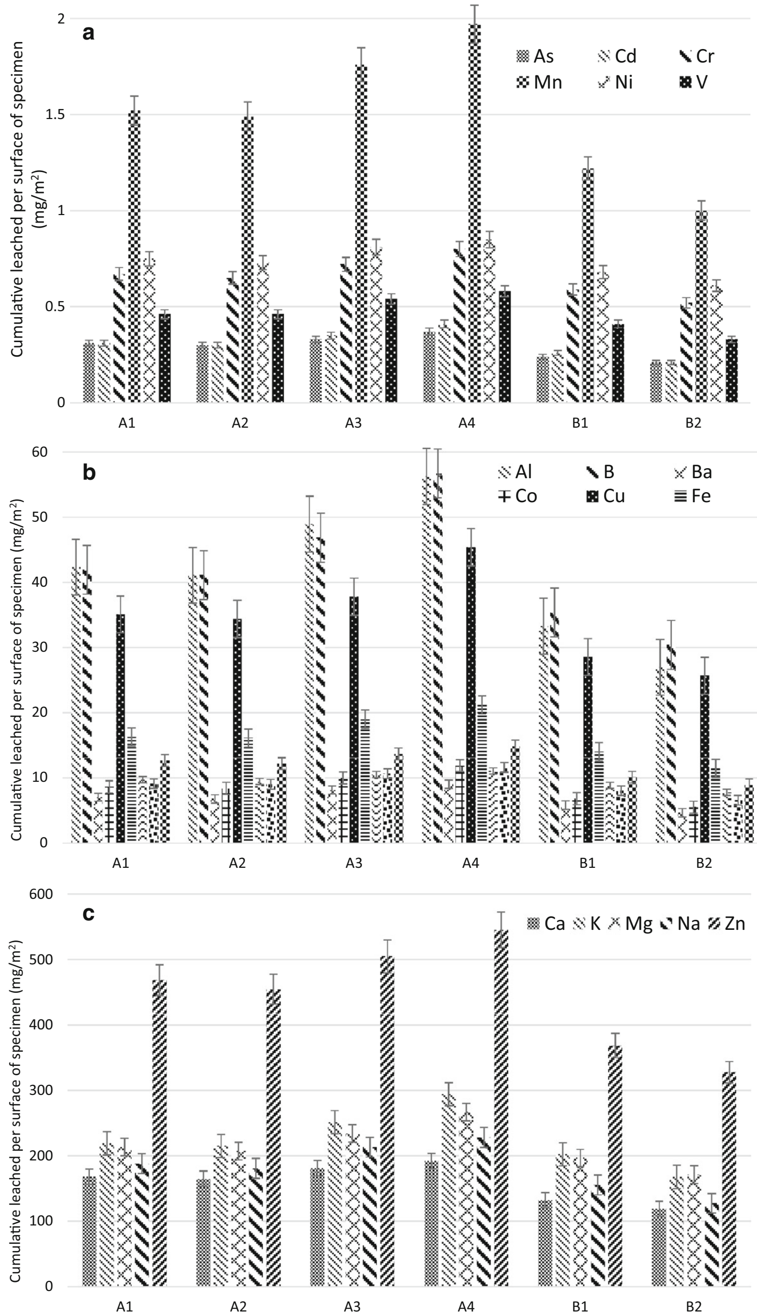


Fig. 9 Changes in (**a**) Electric Conductivity and (**b**) pH of leachants at different stages of DSLT for rubberised asphalt specimens


a
As
Cd
Cr
Mn
Ni
V
Cumulative leached per surface of specimen (mg/m²)
A1
A2
A3
A4
B1
B2
b
Al
B
Ba
Co
Cu
Fe
Cumulative leached per surface of specimen (mg/m²)
A1
A2
A3
A4
B1
B2
c
Ca
K
Mg
Na
Zn
Cumulative leached per surface of specimen (mg/m²)
A1
A2
A3
A4
B1
B2

◀**Fig. 10** Cumulative release of elements from unit surface of rubberized asphalt specimens. **a** Elements with release concentration < 6 mg/m^2, **b** Elements with release concentration between 6 and 60 mg/m^2, **c** Elements with release concentration > 60 mg/m^2

samples that stayed in contact with leachants for 28 days. The observed pH was associated to B1 mix design with a value of 9.35 on day 64 of DSLT. A deeper look at data in Fig. 9b reveals that leachants from B1 and B2 mix designs had lower pH than others at all stages of DSLT except day 36, where they showed the highest pH. This observation may be due to the slower release of ions from B1 and B2 mix designs, which reached their maximum on day 64 and 36, respectively. This phenomenon was observed in Fig. 1a where maximum electric conductivity of leachants from B1 and B2 mix designs occurred on day 64 and 36. Maximum pH values for A-mix designs occurring at earlier stages of DSLT were also in line with their maximum electric conductivity occurrence at earlier stages (Fig. 9a).

4.3.2 Release of heavy metals and trace elements

Samples from different stages of DSLT for different rubberised asphalt mix designs in this study were analysed using ICP-OES. The mean concentrations of heavy metals and trace elements including Al, As, B, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Mo, Na, Ni, Pb, Sb, Se, Si, Tl, V and Zn, were quantified. The cumulative release of elements per unit surface of samples during DSLT (from start to day 64) are presented in Fig. 10. Figure 10a shows the release of elements with concentrations less than 6 mg/m^2. Elements with released concentrations between 6 and 60 mg/m^2 are shown in Fig. 10b. Figure 10c presents the leached elements with highest concentrations during DSLT of different rubberised asphalts. As illustrated in Fig. 10a, the A4 mix design released the highest As (0.37 mg/m^2), Cd (0.41 mg/m^2), Cr (0.8 mg/m^2), Mn (1.97 mg/m^2), Ni (0.84 mg/m^2) and V (0.58 mg/m^2). The B2 mix design showed the lowest release of elements into the leachant, followed by B1, A2, A1 and A3 mix designs. The same trend was observed for elements with releases per unit surface area between 6 and 60 mg/m^2. The overall release per surface area of mix designs for Al, B, Ba, Co, Cu, Fe, Mo, Pb and Si elements were as follows: A4 > A3 > A1 > A2 > B1 > B2. As the data shows, the release of elements from B-mix designs were generally lower than A-mix designs. This observation may be due to a lower content of crumb rubber in B1 and B2 mix designs, which reduces the potential of elemental leaching into the leachant. As Fig. 10c illustrates, the minimum mean released Ca (118.71 mg/m^2), K (167.64 mg/m^2), Mg (171.14 mg/m^2), Na (127.39 mg/m^2) and Zn (327.86 mg/m^2) elements during DSLT were associated with the B2 mix design. For all mix designs, the highest released element per unit surface area of samples were Zn, K, Mg, Na and Ca with concentrations higher than 100 mg/m^2. The observed results were in line with previous studies which reported Zn as the highest leached elements from crumb rubber granulates used in artificial turfs [37–40]. The concentrations of all elements under investigation in this study were lower than detection limit of the ICP-OES instrument which satisfied the second stage of quality check of the experiments and guaranteed no contamination of experimental setup. There is a lack of limitations set by the regulatory bodies on maximum released heavy metals and trace elements, per unit surface area of samples, from DSLT tests in mg/m^2 unit. Therefore, Dutch Soil Quality Decree (SQD) (2006/0496/NL) [41] values were selected to compare the cumulative released elements from rubberised asphalt with the thresholds. The SQD imposes, among other, conditions on the use building materials on or in the ground or in surface water [41]. It has set 260, 1500, 3.8, 120, 60, 98, 1.4, 400, 144, 81, 4.8, 50, 320, 800 mg/m^2 as threshold for As, Ba, Cd, Cr, Co, Cu, Hg, Pb, Mo, Ni, Se, Sn, V and Zn, respectively. As Fig. 10 shows, the cumulative release of elements under investigation in this study from unit surface of all rubberise asphalts were lower than the thresholds set by the Dutch SQD.

5 Conclusions

This paper investigated the mechanical performance of a newly developed material and analyses the leaching and potential hazardousness and environmental impact of six rubberised mixtures.

The compressive tests revealed the similarity in the loading behaviour of the mixtures to playgrounds or rubber composite materials able to considerably reduce the injuries. Also, the change of binder can strongly affect the material's behaviour under stress, even when a constant amount of rubber is used.

The A-mixes have higher elasticity and provide the necessary impact attenuation properties. A3, A2 and B2 mixes are less resistant, while the SBS-binder mix shows the best durability and cohesive properties, while mix B1 records a better strength but does not comply with the injury reduction performances. The results from the compressive test showed that the A-mixes have the best elastic and absorbing behaviour, especially the one made with an SBS-modified emulsion. Altogether, mix A4 has a low strength but high elasticity, essential for the impact attenuating behaviour. The use of A4 also brings in some energy-saving and decreased externalities during the production procedure.

The results of DSLT revealed that the B-mix asphalt specimens released lower concentrations of heavy metal and trace elements into the leachants, and A-mix specimens released the heavy metals and trace element ions at earlier stages of the DSLT compared to B-mix design specimens. During DSLT, the cumulative release of all elements under investigation in the present study were lower than maximum thresholds set by Dutch Soil Quality Decree (SQD). The pH of the leachant from DSLT constantly increased during 64 days of the experiments, ranging between 7.2 and 9.3. The electric conductivity of the leachant ranged between 550 and 1080 mS/cm during all stages of DSLT.

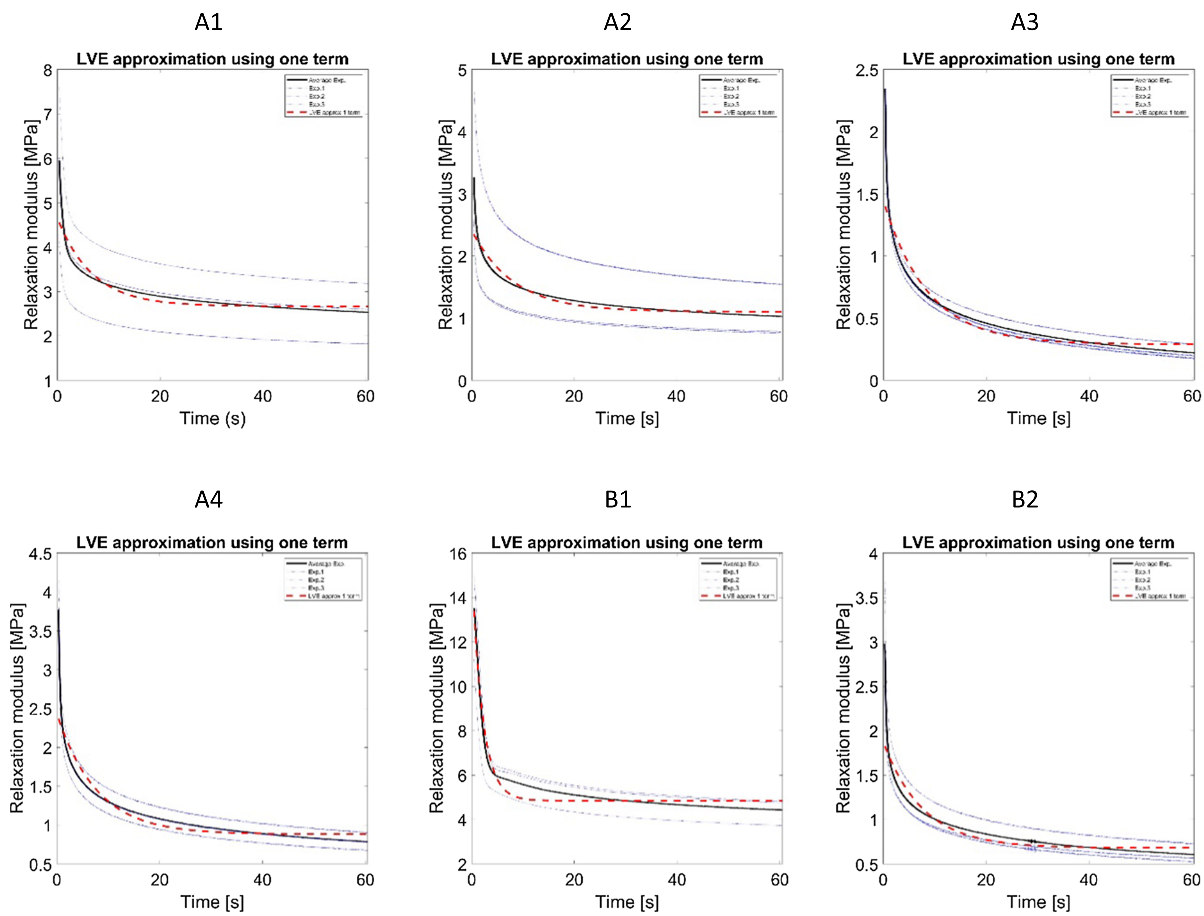


Fig. 11 Experimental relaxation test and fitted SLS/LVE model using 1 term

The A4 mix is the most promising mixture regarding the injury reduction performances and the environmental impact as it uses a cold binder thus, the production process is considerably limiting the externalities. B2 has low leaching values but do not comply with the durability and resistance needs. B1 mechanical and leaching performances are acceptable however the injury reduction one is not appropriate for the aim application. At valuable mechanical and leaching performance, A1 seems an interesting alternative but barely comply with the injury reduction requirements.

Therefore, the early release of leachant observed for the A-mix, especially A4, suggests the possibility of handling the leaching with several solutions, including rubber coating treatment or water washing before their incorporation into the mix to prevent their leaching while enabling very high injury reduction performances.

Acknowledgements The authors are grateful to the crumb rubber (through the participation of Svensk Däckåtervinning AB) and bituminous binders' suppliers used during this study and to Ing. Filippo Venturucci for his contribution to the testing phase.

Authors Contribution CM, AL: Conceptualisation; CM, AL: Data curation; CM, AL: Formal analysis; CS, SC: Funding acquisition; CM, AL: Investigation; CM, AL, SK, SC: Methodology; CM, AL, CS, SK, SC: Validation; CS, SK, SC: Supervision; CM, AL: Writing—original draft; CM, AL, SK, CS, SC: Writing—review & editing. All authors have read and agreed to the submitted version of the manuscript.

Funding Open access funding provided by Alma Mater Studiorum - Università di Bologna within the CRUI-CARE Agreement. This research was funded by the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie Grant Agreement No 765057 through SAFERUP! Project.

Declarations

Conflicts of Interest The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

Appendix

Table 3 summarises the Relaxation Modulus at the beginning of the application of the load G_0 and G_{inf} Relaxation Modulus after a long time, which is lower in value for elastic behaviour due to elastic stress redistribution values of the six mixes. The mixture showing the highest G_0 is B1 followed by A1, A4, A2, B2 and A3. The highest G_{inf} is observed for B1 followed by A1, A2, A4, B2 and A3. B1 has the highest standard deviation while A3 has the lowest. These values confirm the repeatability and liability of the test done on the samples.

Additionally, the experimental relaxation modulus of each mixture was calculated and fitted to the Standard linear viscoelastic model (SLS or LVE). Figure 11 shows the results. These relaxation behaviours validate the viscoelastic approximation used for the calculations for the compressive testing procedure. The maximum relaxation modulus value varies from 14 MPa (B1) to 3 MPa (A3).

References

1. World Health Organization (2021) Road traffic injuries. https://www.who.int/news-room/fact-sheets/detail/road-traffic-injuries. Accessed 22 Jan 2022
2. European Conference of Ministers of Transport (2000) Safety in road traffic for vulnerable users. OECD
3. Kraus S, Koch N (2021) Provisional COVID-19 infrastructure induces large, rapid increases in cycling. PNAS. https://doi.org/10.1073/pnas.2024399118
4. Ardefors F, Roupé J (2020) The road to sustainability. White paper 2020–2030. The Swedish Tyre Industry Association
5. Fédération Internationale de Football Association (FIFA) (2015) Fédération internationale de football association (FIFA) Quality programme for football turf handbook of test methods.
6. Huang TJ, Chang LT (2009) Design and evaluation of shock-absorbing rubber tile for playground safety. Mater Des 30:3819–3823. https://doi.org/10.1016/j.matdes.2008.12.024

7. Li X, Kleiven S (2018) Improved safety standards are needed to better protect younger children at playgrounds. Sci Rep. https://doi.org/10.1038/s41598-018-33393-z
8. Wallqvist V, Kjell G, Cupina E et al (2017) New functional pavements for pedestrians and cyclists. Accid Anal Prev 105:52–63. https://doi.org/10.1016/j.aap.2016.04.032
9. Makoundou C, Johansson K, Wallqvist V, Sangiorgi C (2021) Functionalization of crumb rubber surface for the incorporation into asphalt layers of reduced stiffness: an overview of existing treatment approaches. Recycling 6:19. https://doi.org/10.3390/recycling6010019
10. Kraft L, Rogers P, Brandels A, et al (2015) Experimental rubber chip concrete mixes for shock absorbent bike lane pavements. 57 Congresso Brasileiro do Concreto - CBC2015, Bonito, Brasil
11. Makoundou C, Sangiorgi C, Johansson K, Wallqvist V (2021) Development of functional rubber-based impact-absorbing pavements for cyclist and pedestrian injury reduction. Sustainability 13:11283. https://doi.org/10.3390/su132011283
12. Wallqvist V, Kraft L (2015) Prototype bike lanes -placement practices and properties. 57° Congresso Brasileiro do Concreto -CBC2015, Bonito, Brasil
13. Makoundou C, Sangiorgi C (2022) Influence of Freeze–Thaw Cycles on the Mechanical Properties of Highly Rubberised Asphalt Mixtures Made with Warm and Cold Asphalt Binders Materials 15(7):2701. https://doi.org/10.3390/ma15072701
14. Siverio Lima MS, Hajibabaei M, Hesarkazzazi S et al (2020) Environmental potentials of asphalt materials applied to urban roads: case study of the city of münster. Sustainability 12:6113. https://doi.org/10.3390/su12156113
15. Siverio Lima MS, Hajibabaei M, Hesarkazzazi S et al (2021) Determining the environmental potentials of urban pavements by applying the cradle-to-cradle LCA approach for a road network of a midscale German city. Sustainability 13:12487. https://doi.org/10.3390/su132212487
16. Fathollahi A, Coupe SJ (2021) Life cycle assessment (LCA) and life cycle costing (LCC) of road drainage systems for sustainability evaluation: quantifying the contribution of different life cycle phases. Sci Total Environ 776:145937. https://doi.org/10.1016/j.scitotenv.2021.145937
17. Cheng H, Hu Y, Reinhard M (2014) Environmental and health impacts of artificial turf: a review. Environ Sci Technol 48:2114–2129. https://doi.org/10.1021/es4044193
18. Rhodes EP, Ren Z, Mays DC (2012) Zinc Leaching from tire crumb rubber. Environ Sci Technol 46:12856–12863. https://doi.org/10.1021/es3024379
19. Celeiro M, Dagnac T, Llompart M (2018) Determination of priority and other hazardous substances in football fields of synthetic turf by gas chromatography-mass spectrometry: A health and environmental concern. Chemosphere 195:201–211. https://doi.org/10.1016/j.chemosphere.2017.12.063
20. Llompart M, Sanchez-Prado L, Pablo Lamas J et al (2013) Hazardous organic chemicals in rubber recycled tire playgrounds and pavers. Chemosphere 90:423–431. https://doi.org/10.1016/j.chemosphere.2012.07.053
21. Menichini E, Abate V, Attias L et al (2011) Artificial-turf playing fields: Contents of metals, PAHs, PCBs, PCDDs and PCDFs, inhalation exposure to PAHs and related preliminary risk assessment. Sci Total Environ 409:4950–4957. https://doi.org/10.1016/j.scitotenv.2011.07.042
22. Ruffino B, Fiore S, Zanetti MC (2013) Environmental–sanitary risk analysis procedure applied to artificial turf sports fields. Environ Sci Pollut Res 20:4980–4992. https://doi.org/10.1007/s11356-012-1390-2
23. Schneider K, de Hoogd M, Madsen MP et al (2020) ERASSTRI–European risk assessment study on synthetic turf rubber infill–part 1: analysis of infill samples. Sci Total Environ 718:137174. https://doi.org/10.1016/j.scitotenv.2020.137174
24. Tóth G, Hermann T, Da Silva MR, Montanarella L (2016) Heavy metals in agricultural soils of the European Union with implications for food safety. Environ Int 88:299–309. https://doi.org/10.1016/j.envint.2015.12.017
25. Bandow N, Gartiser S, Ilvonen O, Schoknecht U (2018) Evaluation of the impact of construction products on the environment by leaching of possibly hazardous substances. Environ Sci Eur 30:14. https://doi.org/10.1186/s12302-018-0144-2
26. Gupta N, Kluge M, Chadik PA, Townsend TG (2018) Recycled concrete aggregate as road base: leaching constituents and neutralization by soil Interactions and dilution. Waste Manage 72:354–361. https://doi.org/10.1016/j.wasman.2017.11.018
27. Maia MB, De Brito J, Martins IM, Silvestre JD (2018) Toxicity of recycled concrete aggregates: review on leaching tests. Open Constr Build Technol J. https://doi.org/10.2174/1874836801812010187
28. Solouki A, Fathollahi A, Viscomi G et al (2021) Thermally treated waste silt as filler in geopolymer cement. Materials 14:5102. https://doi.org/10.3390/ma14175102
29. Spreadbury CJ, Clavier KA, Lin AM, Townsend TG (2021) A critical analysis of leaching and environmental risk assessment for reclaimed asphalt pavement management. Sci Total Environ 775:145741. https://doi.org/10.1016/j.scitotenv.2021.145741
30. Fathollahi A, Makoundou C, Coupe SJ, Sangiorgi C (2022) Leaching of PAHs from rubber modified asphalt pavements Science of The Total Environment 826153983. https://doi.org/10.1016/j.scitotenv.2022.153983
31. Introduction to the EU Water Framework Directive - Environment - European Commission. https://ec.europa.eu/environment/water/water-framework/info/intro_en.htm. Accessed 29 Jan 2022
32. Genan rubber granulate - choose between 6 different standard fractions. In: Genan. https://www.genan.eu/products/granulate/. Accessed 12 Jul 2020
33. Davies J, Wallace WA, Colton C, et al (2019) The Head Injury Criteria and Future Accident Investigations. In: Proceedings of the International Society of Air Safety Investigators (ISASI) Annual Seminar. The Hague
34. Hutchinson J, Kaiser MJ, Lankarani HM (1998) The head injury criterion (HIC) functional. Appl Math Comput 96:1–16. https://doi.org/10.1016/S0096-3003(97)10106-0
35. IBM SPSS Statistics V21.0
36. Li X, Kleiven S (2018) Improved safety standards are needed to better protect younger children at playgrounds. Sci Rep 8:15061. https://doi.org/10.1038/s41598-018-33393-z

37. Edeskär T (2006) Use of tyre shreds in civil engineering applications: technical and environmental properties. Luleå University of Technology.
38. ETRMA (2011) End of life tyres: a valuable resource with growing potential. European Tyre and Rubber Manufacturers Association, Brussels
39. Twin City Testing Corporation, Minnesota Pollution Control Agency, Waste Tire Management Unit (1990) Waste tires in sub-grade road beds: a report on the environmental study of the use of shredded waste tires for roadway subgrade support. Waste Tire Management Unit, Site Response Section, Groundwater and Solid Waste Division, Minnesota Pollution Control Agency, St. Paul, Minn.
40. Downs LA, Humphrey DN, Katz LE, Rock CA (1996) Water quality effects of using tire chips below the ground water table. University of Maine, Orno
41. (2007) Soil Quality Decree. (2006/0496/NL). https://rwsenvironment.eu/publish/pages/126568/soilqualitydecree_24_275037.pdf