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Feasible use of colliery spoils as subbase layer for low-traffic roads

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Abstract

The aim of this paper is to characterise, both physically and chemically, the colliery spoils of *Pozo San José*, located in the Guadiato Valley (Córdoba, Spain), and evaluate their possible use as unbound materials or as materials bound with lime or cement in the construction of roads with low traffic intensity. The leaching of the material and its effect on the environment was also studied. The results obtained show that the main limiting property of the material is its high organic matter content. However, colliery spoils can be used unbound or bound with 2% of cement as a subbase, thus giving a second life to the colliery spoils of this mining region that currently have no purpose. The use of colliery spoils bound with lime is not recommended, since their mechanical properties are not improved. Finally, two sections are designed for roads with a traffic intensity of up to 24 heavy vehicles / day. This research contributes to the promotion of the new paradigm of the circular economy in the construction sector.

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1. Introduction

The new paradigm of sustainable development and circular economy in the construction and mining sectors involves reducing the consumption of raw materials from the earth's crust, not exhausting their availability for future generations, minimizing the environmental impact derived from the generation of waste, and favouring their incorporation into a productive process. The use of colliery spoils as a secondary raw material in the construction sector contributes to the reduction of the consumption of non-renewable natural resources, such as aggregates, giving a second life to the waste of this extractive activity. It is possible to use this waste in the construction of roads with low traffic intensities, as long as the technical and environmental conditions required by the regulations of different countries are met [1], as has been done with the use of recycled aggregates from construction and demolition wastes [2] [3].

Coal is the second most consumed primary source of energy in the world. Approximately 70% of the coal consumed in the world in 2015 was used to generate electricity. Thus, 40.8% of the electricity produced in 2015 was produced with coal [4]. Coal continues to be, therefore, one of the backbones of the generation of electric power and a basic fuel for the industrialisation and growth of emerging economies. According to the “BP Statistical Review of World Energy, June 2017,” the total coal consumption in 2016 was 3732 million tons, with Asia being the largest producer with 2753.6 million tons. In Spain, the consumption for the year 2016 was 10.6 million tons [5].

Sterile coal is the product of the separation of coal from unusable components. These unusable components originate in the excavation of coal mines and anthracite mines, consisting of the encapsulating rocks in the coal layers (the *mining spoils*), as well as material generated in the processes of coal washing (the *tailings*). These barren wastes are those resulting from the storage of mine waste and the washing of rubble. These materials are normally stored next to coal mines in tailings that fill valleys, change the relief and degrade the landscape. The resulting barren wastes are also known by the name of “colliery spoils” or “minestone” [6] [7].

The European Directive 2006/21/CE of waste management for extractive industries establishes the objectives of recovery and recycling [8]. The member countries have incorporated the European Directive into their regulations, as is the case in Spain. However, at present, colliery spoils continue to accumulate in the areas adjacent to the extraction facilities, a problem that is aggravated when the exploitation activity ceases.

Skarzynska [9] provides the physical, chemical and mechanical characterisation of mine wastes from 13 countries in America, Europe and Asia, showing that the properties of these wastes are similar in the different countries studied and concluding that we can utilise them with appropriate caution. The reuse or recycling of mining waste is not new. The oldest method of reusing mining waste is to adapt the waste to the landscape through reforestation or agricultural management [10]. There are studies on the use of colliery spoils or mining spoils as road embankments [11]. Regarding its geotechnical characteristics, the colliery spoils usually have well-graduated granulometric curves and their dry densities obtained from the normal proctor test are approximately 2 g/cm^3. These types of materials usually lack cohesion, and their internal friction angle ranges between 30 and 40 degrees [12].

The use of colliery spoils mixed with Portland cement has also been studied by several authors [13] [14]. The percentages of cement used usually ranges between 8 and 10%. The Car Park at Gatwick airport in London was paved using cement-bound minestone [15]. In Spain, there are studies in which colliery spoils have been used for the construction of road embankments [16]. The construction of the subbase of a 4.5 km stretch of highway between Oviedo-Campomanes (Asturias, Spain) incorporated 53,000 t of colliery spoils stabilised with 6% of cement [17]. The size of the particles is a limiting factor in the colliery spoils bound with cement; as it increases, the strength of the cement-bound minestone decreases [10].

Other studies have shown that coal waste stored in slag heaps can be used with bituminous emulsions for the construction of layers of roads at ambient temperature and without risk to the environment by the leaching of pollutants. This mixture improves the mechanical properties by increasing the Marshall stability, the tensile strength and the resilient modulus of the material [18]. This same waste in powder form is used as a filler in a hot bituminous mix, which improves its properties compared to reference mixtures, [19]. It has also been used for river embankments [20], dykes and dams [21]. For railway platform embankments, it has been used in two sections of United States railroads and within the London-Brighton commuter line located in Croydon in Surrey, having a total length of 1.1 km. [22]. Filling existing mines with the waste and adapting the waste to the landscape are the oldest uses but are also the least compatible with the new paradigm of the circular economy [23]. However, despite these advances, colliery spoils still have no application in the Guadiato Valley (Andalusia, Spain) due to the lack of studies characterising them and studying their possible uses as secondary materials.

The region of the Guadiato Valley is located in the north of Córdoba (Andalusia, Spain). The most important economic activity since the mid-nineteenth century has been mining, which has led to significant changes in the landscape, with slag heaps that have a great visual impact. The San José mine, an extraction mine that was active from 1960 to 1993, is the longest-operating mining activity of this mining centre, providing a total of more than 3 million tons of anthracite and producing almost the same amount of sterile coal [24].

Globally, the amount of sterile coal mining will increase in the coming years as a result of the worsening conditions of the deposits. The results of this research can help solve this problem both locally and globally.

The aim of this paper is to characterise and evaluate the possibilities of using colliery spoils from the San José mine, located in the region of Guadiato Valley, in the northern region of the province of Córdoba (Spain), as a material for the construction of low traffic-intensity roads, both in its unbound state or bound with either lime or cement. In addition, the environmental impact will also be studied through leaching tests. All these results will be compared with the national reference regulation, which is Spanish general technical specification for road construction (PG-3) [25]. In addition, solutions have been calculated for typical sections of roads with a traffic intensity of less than 24 heavy vehicles / day, which will help to find a use for this type of material and to promote the new paradigm of a circular economy in the construction sector.

2-Material and methods

2.1 Collection and preparation of samples

The material is stored at the *Pozo San José* mine, located in the municipality of Fuenteobejuna, within the mining region of the Guadiato Valley (Córdoba, Spain). Fig. 1 shows an aerial photo of the stockpile.

Fig. 2 shows the collection of colliery spoils, which has an area of 26000 m^{2}, with an average height of 12 m, such that the estimated amount of waste is 624000 tons.

So that the characterisation of the colliery spoils was representative, samples from different places in the collection were taken following the specifications of the UNE-EN 932-1 standard in February of 2017.

There are similar stockpiles at other mines in the area. The reason for choosing the *Pozo San José* is because it is the longest standing mining site in the Guadiato Valley and because the results can be extrapolated to the rest of the mines in the region, as stated by Skarzynska [9]. The amount of colliery spoils accumulated throughout the Guadiato Valley has been estimated at 4.2 million tons.

2.2 Physico-mechanical characterisation

The physico-mechanical characterisation of the material has been carried out in such a way that we can compare the results obtained with what is established in articles 330, 510 and 512 of PG-3 [25]. In PG3, the requirements are established for the materials and for their use in the structural layers of roads in Spain. Table 1 shows the characterisation tests carried out.

Fig. 3 shows the composition of the material: dark shale (60%), soft shale (31%), coal anthracite (8%) and other minor constituents such as diabolised kaolin (1%). The Guadiato coalfield corresponds to a tectonic pit of Westfalian age B (approximately 300 million years old) embedded in Precambrian and Paleozoic rocks. There are two types of coals clearly identified: anthracite and coal. In this case, the colliery spoils studied comes from the anthracitic coal excavation. The results presented in Fig. 3 are in accordance with geological map 879 of the IGME [26], which indicates that the different layers of coal are interspersed with agglomerates of sand and shales are indicated.

2.3 Chemical characterisation

The chemical characterisation (Table 2) was carried out according to the standardized tests that are established in articles 330, 510 and 512 of the General Technical Prescriptions Sheet for road and bridge works of Spain (PG-3), as well as others that have been deemed necessary to be able to discuss the suitability of the material for its use on roads [25].

2.4 Mineralogical characterisation

The material was characterised by X-ray diffraction (XRD) techniques, differential thermal analysis (DTA) and thermogravimetric analysis (TGA), in addition to energy dispersive X-ray analysis (EDX). The material was analysed using a Bruker D8 Discover A25 instrument with CuKα radiation. All diffraction patterns were obtained by scanning the goniometer from 3° to 70 ° (2θ) at a rate of 0.006 θ min-1. The thermogravimetric analysis was performed in a Setaram Setsys Evolution 16/18 apparatus at a heating rate of 5 ° min-1. The working temperature ranged from ambient temperature to approximately 1000 °C.

2.5 Leaching test

From an environmental point of view, to study the possible leaching of the colliery spoils and to observe if their use is potentially dangerous for the environment, the UNE-EN 12457-3: 2002 standard was used. A dry mass of 175 grams of the fine and coarse fractions that were previously ground to guarantee sizes smaller than 2 mm was analysed.

This test aims to simulate two exposure scenarios, short- and long-term. To simulate the short-term scenario, sufficient deionised water to achieve a liquid / solid ratio (L/S) of 2 L/kg was added, the sample was flipped for 6 hours at a speed between 5–10 rpm, and then was filtered with a 0.45 μm filter. To simulate the long-term scenario, deionised water was added to the previous sample until obtaining an L/S ratio of 10 L/kg. The sample was flipped for 18 hours and then filtered with a 0.45 μm filter.

The leaching tests were conducted in duplicate. The quantities of several major and trace elements were determined in the laboratory using inductively coupled plasma mass spectrometry (ICP-MS). The ICP-MS (model Perkin-Elmer ELAN DRC-e) was equipped with a sample introduction system with a dilutor and was capable of argon plasma ionisation and quadruple ion detection. The machine had a dynamic reaction cell (DRC) cell for interference suppression.

The results were compared with the waste-acceptance criteria for landfilling (EU Council, 2003), stated in Annex 2 of the 2003/33/CE Council Decision (based on the 1999/31/EC Directive) [27] .

To guarantee that the high organic matter content of colliery spoils will not be a hazard to the environment, the total organic carbon (TOC), total inorganic carbon (TIC) and total carbon (TC) were determined according to the standard UNE-EN 15936: 2012 (equivalent to UNE EN 13137: 2002).

Table 3 summarises the potential pollutants and the limits for the material to be classified as inert, non-hazardous and hazardous.

2.6 Tests of colliery spoils bound with cement

To improve the physico-mechanical properties of the colliery spoils, its use when bound with cement was studied. The cement used for the manufacture of test samples was CEM II / B-L 32.5 N (UNE-EN 197-1: 2011).

Table 4 shows the tests that were performed on the colliery spoils mixed with cement. The materials mixed with cement must meet the technical specifications of article 513 and article 512 of PG-3 [25] to be used as a soil-cement or stabilised soil, respectively. According to PG-3 [25], the soil-cement can be used in base and subbase structural layers.

Stabilised soil, according to PG-3 [25], is the mixture of a soil with a binder (lime or cement) to increase its dimensional stability and bearing capacity. Stabilised soil can be used in subbase and embankment layers. According to its final mechanical properties, three types of stabilised soils can be used, from lowest to highest bearing capacity: S-EST 1 (bound with lime or cement), S-EST 2 (bound with lime or cement) and S-EST 3 (only bound with cement).

2.7 Tests of colliery spoils bound with lime

In the same way, the effect of the incorporation of different percentages of lime in the physical-mechanical properties of colliery spoils was studied, although no references have been found regarding the use of colliery spoils with lime.

The lime used was an air and calcium lime with a magnesium content of less than 5%, CL 90-S type, complying with the UNE 80502: 2003 standard "live or hydrated limes used in the improvement and/or stabilization of soils". This is also the lime recommended in Section 512.2.2. of the PG-3 [25] . Table 4 lists the tests carried out with lime, which are the same as those carried out with cement.

2.8 Description of sections

Two sections will be proposed: the first section made with unbound colliery spoils and the other section made with colliery spoils bound with a selected percentage of lime or cement. To calculate these sections, Everstress software has been used, which has been developed by the Washington State Department of Transportation from WESLEA layered elastic analysis software (provided by the Waterways Experiment Station, U.S. Army Corps of Engineers) [28]. The interface between the different layers will be specified as fully bonded.

The layered elastic analysis is the tool most often used to calculate flexible-pavement response to traffic loads. This is mainly due to its simplicity and the fact that pavement engineers have used it since 1940 [29]. Burmister et al [30] developed a closed-form solution for a two-layered linearly elastic half-space problem, with the advances in computer technology, the theory has been extended to deal with multilayer systems. To carry out this analysis, the material characteristics are needed, such as the Poisson coefficient and the resilient modulus.

To simulate the traffic loads, a semi-axle vehicle with double tyres was considered for this analysis. The distance between the tyres is 37.5 cm. The contact pressure is 0.8 MPa and the wheel print radius is 11.35 cm. For this configuration, each tyre was loaded with 37.376 KN. This traffic load is proposed for the guidelines for road surfacing in Andalusian highways [31]

The design of traffic intensity will be a maximum of 24 heavy vehicles per day, which is considered in the Spanish Instruction of Highway as traffic category T42 [32].

Taking into account a service life of 10 years and an annual traffic growth rate of 2%, the number of equivalent axles calculated according to the guidelines for road surfacing in Andalusian highways is 60000 [31]. The method used is similar to ESAL (equivalent single axis load) [33,34] widely used in the literature, which defines the relationship between the damage caused by the passage of a standard axis on the same pavement.

To analyse the pavement layers failure, the following critical parameters have been considered:

- The maximum tensile stress (σ_r) in cement-treated materials
- The unitary vertical deformation in the upper face (ε_z) in the subbase

The guidelines for road surfacing in Andalusian highway [31] proposes the following fatigue laws for cement treated materials (1) and unbound granular materials (2), which determines the admissible number of load applications (N). This value of N is that obtained as equivalent single axle load of the service life of road.

$$\theta_r\ (MPa) = 0{,}72 \cdot (1 - 0{,}065 \cdot \log(N)) \qquad (1)$$

$$\varepsilon_z(-) = 2.16 \cdot 10^{-2} \cdot N^{-0.28} \qquad (2)$$

The design of sections made with colliery spoils will be useful to promote the use of these materials in low-traffic roads construction, such as Barbudo et al. [35] made with the recycled aggregates from construction and demolition waste. These designed pavements will be used in a future study to build an experimental road section in order to know the performance of the colliery spoils under real traffic conditions.

3-Results and discussion

3.1 Characterisation of colliery spoils

3.1.1 Particle size distribution

Fig. 4 shows the particle size distribution curves of colliery spoils before and after compaction. Colliery spoils has a continuous granulometry, with a quantity of fines (<2 mm) of approximately 22%.

To study the susceptibility to compaction, after compacting the material, another particle size distribution test was carried out. The effective diameter (D_{10} is the particle diameter that corresponds to the 10% interval) of the initial granulometry has a value of 0.65 mm, while in the granulometry after compaction, it has a value of 0.40 mm. The value of the uniformity coefficient ($C_u=D_{60}/D_{10}$) remains almost constant before and after the compaction, with values of 13.26 and 13.75, respectively, which indicates that the material has an extended granulometry. The coefficient of curvature or concavity ($C_c=D_{30}^2/(D_{60}*D_{10})$) has a value of 1.71 before compaction and 1.60 after compaction, indicating a well-graduated soil. In terms of engineering, soils with that have extended granulometry and are well-graduated have better performance, so it is highly recommended to use them as a subbase or base structural layers in road construction.

3.1.2 Physico-mechanical properties

The Flakiness Index (FI) yielded a result of 30% that is in the same order as that obtained by Cadierno [7]. This value is lower than the limit of 35 imposed by most regulations for use as subbase or base material [25], so this is not a limiting property.

Density and water absorption results for the coarse fraction (4 / 31.5 mm) and for the fine fraction (4 / 0.063 mm) are summarised in Table 5. The density values are similar to those presented by Cañibano [17] and are also similar to the maximum obtained by Holubec in the U.S.A. and in the United Kingdom [12].

To determine the plasticity of the colliery spoils, Atterberg Limit tests were conducted, which measured a liquid limit (LL) value of 26 and a plastic limit (PL) value of 19; and the value of plasticity index (PI) was of 7. This could be considered low plasticity, similar to that found by Fernández et al. [36] in the colliery spoils of the northern region of Spain.

Regarding the behaviour tests, the modified Proctor test, which relates the dry density and the moisture content, was carried out, providing the results shown in Fig. 5 A, which yielded a maximum density of 2.18 g/cm^3 at the optimum moisture content of 7.2%. Compared to Cañibano (1.98 g/cm$^{3;}$ 13.5%), the dry density was higher at a lower optimum moisture content [17]. Fernández et al. [36] indicates that these types of waste have to reach a dry density of at least 2.08 g/cm^3, in accordance with the experimental results [37].

The clean coefficient was 1.95%. The Angeles test was also carried out to determine the resistance to fragmentation, with a result of 30%.

To determine the bearing capacity, the CBR test was carried out; the material was compacted to the optimum humidity obtained from the modified Proctor, and the CBR test was carried out under 4 days soaked conditions and an overload of 4.5 kg, obtaining the results shown in Fig. 5 B (for compaction energy values of 0.658 J/cm^3, 1.316 J/cm^3, and 2.632 J/cm^3, which match the dry densities of 1.94, 2.1, and 2.17 g/cm^3, respectively). The CBR values presented by Vadillo Fernández [37] reach a maximum value of 29 in the sterile slag heaps of the Centre-South of Spain and those of Centre-North, similar to those obtained from the colliery spoils in our study (27). The highest observed swelling, after the 4-day soaked conditions, was 0.85%, which is negligible from an engineering point of view.

3.1.3 Chemical properties

The results of the chemical characterisation are shown in Table 6. Regarding organic matter, the potassium permanganate test, which is recommended by PG-3 [25], showed a result of 1.92%. The test, which determines the content of organic matter with potassium permanganate, may not be entirely appropriate for colliery spoils since the test is based on an oxidant mass balance, which is reduced (depleted) by reacting with other oxidizable species. Mineral coal is not oxidizable by this method.

The loss by calcination at 550 °C and 950 °C has been used in waste with a high content of organic matter, such as sludge from the paper industry [38]. The calcination loss shows values of 8.38% and 9.30% for 550 °C and 950 °C, respectively, which are well above the value obtained by the potassium permanganate test. The value obtained at 950 °C is very similar to that obtained by X-ray dispersive energy (EDX) (9.94%). The results corroborate the fact that the use of potassium permanganate is not adequate to determine the content of organic matter in this type of waste. The values obtained with these techniques are higher because, in addition to the organic matter, the mineral carbon content must be taken into account, which, according to the value obtained in the composition test, is approximately 8% (see Fig. 3).

A more detailed study of the mineral compounds is necessary, using TGA and DTA. The TGA and DTA results are shown in Fig. 6. The first weight loss, up to approximately 100 °C, corresponds to the humidity (water). At approximately 450 °C, a new weight loss is observed that may correspond to the calcination of the coal (450-500 °C), which is in accordance with the presence of the carbon detected in the analysis energy dispersive of X-ray (EDX). Approximately 460 °C appears an exhotermal peaks which are related to the combustion of the volatile matter and the fixed carbon. This result is in accordance with the others as indicated by Sun et al and Taha et al. [39,40]

For the use of colliery spoils unbound or bound with lime or cement in road structural layers, the contents of soluble salts, gypsums and sulphur compounds are limited by PG-3 [25] to ensure the stability of the sections and avoid the appearance of related pathologies such as expansion and decrease in bearing capacity. The presence of sulphates requires the use of sulphur-resistant cement in soils treated with cement. Four types of tests have been carried out: gypsum content, performed with the NLT 115 standard to comply with PG-3 (Section 330.4.4.3), in which a value of less than 0.01% was obtained; water-soluble salts according to UNE 103205: 2006, in which a value of 0.149% was obtained; total sulphur compounds, to be compared with that indicated in articles 510 and 513 of PG-3 (Section 510.2.2.2 and Section 513.2.3.2); and acid-soluble sulphates, to be compared with the value that is established in article 513 of PG3 (Section 513.2.3.2) [25]. Additionally, to have better control of the problem of sulphates by not using sulphur-resistant cement, the content of water-soluble sulphates was also analysed. The values obtained in these last three tests were less than 0.01%, as seen in Table 6. Based on the sulphate and gypsum results, the recommendation of Ferreras [7] to use sulphur-resistant cement would not be necessary for the tested colliery spoils in this study.

The chemical composition, determined by EDX and expressed as elements, is presented in Table 7. The predominant elements are silicon (Si) and aluminium (Al), while the content of titanium (Ti) is very low.

The analysis by X-ray diffraction patterns (XRD), shown in Fig. 7, indicates that in the colliery spoils, the majority phase was quartz (SiO_2) [33-1161] [41]. In addition, the following phases can be identified in a much smaller proportion: anatase (TiO_2) [21-1272] [41], siderite ($FeCO_3$) [12-0531] [41], illite ($KAl_2Si_3AlO_{10}(OH)_2$) [02-0056] [41], zeolite (Na_2 $7Al_5$ $7Si_{10}$ $3O_{32}$ $12H_2O$) [34-0524] [41], muscovite (H_4K_2 $(Al,Fe)_6Si_6O_{24}$)[03-0849] [41], nontronite ($(Fe,Al)(Si,Al)_2O_5(OH)H_2O$) [02-0027] [41], kaolinite ($Al_2Si_2O_5(OH)_4$) [29-1488] [41], potassium magnesium aluminium silicate hydroxide ($(K(Mg, Al)_{2.04}$ $(Si_{3.34}Al_{0.66})O_{10}(OH)_2$) [40-0020] [41], calcium and aluminium oxide hydrate ($CaAl_2O_4$ $10H_2O$) [12-0408] [41]. The result obtained by X-ray diffraction patterns is very similar to the one found by Sun et al. [39].

3.1.4 Environmental performance

Table 8 shows the concentrations of leached heavy metals (mg / kg) for the steps L / S = 2 and L / S = 10. The values obtained in both steps were below the limits required for waste to be classified as INERT according to the waste acceptance criteria for landfilling, which is stated in Annex 2 of the 2003/33/CE Council Decision; therefore, the use of colliery spoils does not pose a risk to the environment.

According to the EU landfill legislation in order to deposit solid wastes an inert landfill, total organic carbon (TOC) of the colliery spoils should not exceed a value 30 g/kg .The value of total carbon (TC) , total organic carbon (TOC) and total inorganic carbon (TIC) was of 43.2 g/kg, 5.5 g/kg and 37.7 g/kg respectively, which shows that the high organic matter content of colliery spoils will not be a hazard to the environment.

3.2 Use as unbound materials for subbase and embarkment materials

Table 9 shows the results obtained and the material classification according to the PG-3 [25] for its use on roads. In those regulations, five classes of materials are established, which are denominated as follows from higher to lower quality: selected soils (SS), appropriate soils (AS), tolerable soils (TS), marginal soils (MS) and inadequate soils (IS). The SS and AS can be used as a subbase, and the SS, AS and TS as embankments.

Colliery spoils fulfil all the aspects established in the PG-3 [25] as selected soils (SS) except for the content of organic matter calculated according to the potassium permanganate method (1.92%), so colliery spoils is classified as a tolerable soil (TS).

Based on these results, the use of colliery spoils as embankment materials would be possible, although this use would generate little added value to this waste.

3.3 Use as well-graded crushed rock and well-graded gravel for base materials

To analyse if colliery spoils can be used as well-graded crushed rock (WGCR) or well-graded gravel (WGG), the characterisation results were compared with the limits established in Art. 510 of the PG-3 [25] for their use as base materials (Table 10 and 11).

As has been commented upon previously, colliery spoils have a certain plasticity (PI = 6.26), so the PG-3 [25] does not recommend their use as well-graded crushed rock since it should be free of plasticity. However, the material can be classified as well-graded gravel since the plasticity is close to the limits established for this type of material [31]. Tables 10 and 11 show the limits established by PG-3 so that the material can be used as untreated well-graded gravel in the construction of low traffic-intensity road bases.

3.4 Use of colliery spoils bound with cement or lime.

To study the influence of the incorporation of lime or cement on the mechanical properties and bearing capacity, colliery spoils were mixed with different percentages of lime or cement and the following tests were carried out: simple compressive strength (UNE-EN 13286-41: 2006), CBR Index (UNE-EN 103502: 1995), dry density (UNE-EN 103501: 1994) and splitting tensile strength (UNE-EN 123960-6: 2001). The moisture content used for the manufacture of the specimens was that obtained in the modified Proctor test. Fig. 8 shows the relationship between the percentage of lime or cement and the simple compressive strength of the specimens.

The use of lime did not improve the mechanical properties of the material, and even from 8%, it deteriorated them with respect to those used without lime.

In the case of cement, as the percentage of cement increased, the compressive strength increased, obtaining a value close to the 2.5 MPa required by Art. 513 of the PG-3 [25] for a cement-treated soil for road base layers when 14 or 16% of cement was added, which is a very high value and unviable from the technical, economic and environmental points of view. Fig. 9 shows the simple compressive strength values of the samples mixed with lime or cement with respect to the unbound colliery spoils, observing that it is possible to adjust a lineal regression to the points generated in both the cement and lime tests. Ketlle [42] found values of 6.2 MPa and 1.95 MPa for the simple compressive strength test at the age of 7 days using colliery spoils bound with 10 an 5% of cement respectively. These values were greater than those shown in this study. In the case of the straight line representing the colliery spoil mixed with cement (Fig. 9), a linear relationship was observed with a greater slope than was found for colliery spoils mixed with lime. González-Cañibano [17] used a mining waste with 6% of cement in the subbase of a highway, although not providing the value of the simple compressive strength obtained.

In Spain there are studies [17,43] that have confirmed the findings in other European countries, such as France and the United Kingdom [44], on using colliery spoils as cement-treated soil for road subbases. The percentage of cement commonly used in these studies was between 5 and 6%, although these authors did not indicate the value of the simple compressive strength obtained.

At the same time, to know the evolution of the bearing capacity of the material, the CBR index was calculated using the same percentages of lime or cement as was done for the simple compressive strength test. Fig. 10 shows a linear evolution of the CBR index with the incorporation of lime or cement, with the slope of the straight line adjusted to the mixture with cement being much higher than that of the straight line representing the mixture with lime. The regression coefficients were close to one, which indicates the goodness of the fit of the regression equation. Fig. 11 shows the evolution of the dry density of the specimens used to calculate the CBR index. In the case of lime, there was a decrease in the density due to the drying effect of the material, and therefore, to the modification of the optimum compaction moisture of the modified Proctor test.

To study the evolution over time of the mechanical properties, test samples were prepared containing colliery spoils with 5% of lime or 12% of cement. The tests of compressive strength and splitting tensile strength were carried out at four ages: 3, 7, 28, and 90 days. The results are shown in Fig. 12. The simple compressive strength of the samples mixed with cement followed a normal evolution of the base-cement materials, reaching over 2.5 MPa at 28 days and 3.2 MPa at 90 days, so that the interaction of colliery spoils and cement was acceptable. In the case of lime, a favourable evolution of the simple compressive strength with time was not observed because the values were relatively low. The evolution of the splitting tensile strength was similar to that of simple compressive strength.

To determine if the colliery spoils can be used as a soil-cement or as a soil stabilised with lime or cement, the specifications of Art. 513 and 512 of the PG-3 [25] were taken into account. Table 12 shows the requirements of a material for use as a soil-cement in the base layer of roads. It can be observed that all the requirements are satisfied, except for the simple compression strength, which may be related to the high content of organic matter. It is well known that the content of organic matter delay cement setting, in addition to an apparent decrease in the mechanical performance [45]. This use as soil-cement is not recommended (to use on the structural base layer), because of the low compressive strength achieved despite using high percentages of cement.

Table 13 shows the requirements for a stabilised soil with lime or cement for its use in the subbase layer. The particle size distribution of the colliery spoils did not have enough fines (< 0.063 mm) so that the mixture with lime was not the best option. The use of colliery spoils bound with lime was discarded for two reasons: i) not to satisfy the particle size distribution requirements specified in PG-3 and ii) experimentally demonstrating that the incorporation of lime did not improve the mechanical properties of the material, despite the fact that the material has a certain plasticity [25].

The colliery spoils bound with 2% of cement satisfied all the requirement that are specified in PG-3 [25], to be used as stabilised soil (S-EST1), although the requirements to be used as S-EST2 and S-EST3 are not satisfied because of the organic matter content (see Table 13). No minimum compression strength is necessary for stabilised soils S-EST1 and S-EST2 in subbase layers for road construction.

3.5 Pavement design

Fig. 13 and Fig. 14 show two feasible pavement sections made with colliery spoils in the subbase layer. The first option includes unbound colliery spoils and the other includes colliery spoils bound with 2% of cement as a stabilised soil. Stabilisation with cement aims to reduce the thickness of the subbase layer and maximise the utilisation of colliery spoils.

From an environmental point of view, the use of a cold-mix asphalt (CMA) is the best solutions for the surface layer in low traffic road. CMA is composed of aggregates and bituminous emulsion that does not require pre-heating of the components, such that its storage, spreading and compaction is carried out at environmental temperature. This technique is the least polluting, compared to hot, warn and semi-warn asphalt mixtures, for which 8.1, 6.4 and 3.2 of fuel/ton are needed respectively. The fuel consumption in CMA is minimal, and therefore the CO_2 emissions of CMA are the lowest [46]. However, colliery spoils can be used as a subbase layer in construction solutions with all type of bituminous mixture as a surface layer.

The two sections have four layers: Cold-Mix Asphalt (Surface); Granular base (Base) ; Subbase unbound or bound with cement (Subbase) and Subgrade.

The value of resilient modulus for cold-mix asphalt was 1500 MPa, and a value of Poisson's ratio of 0.35 was used. These values are recommended in the Guidelines for road surfacing in Andalusian highway [31]. Furthermore, these values are similar to those used by Serfass et al. [47]. The value adopted for granular base was of 375 MPa and a Poisson's ratio of 0.35 also recommended in the Guidelines for road surfacing in Andalusian highway [31]. These values are similar to those found by Tavira et al. through back-calculation under real traffic and weather conditions in an experimental section made with recycled aggregates from construction and demolition waste [3] .

For the subbase layer, the resilient modulus value was obtained using Eq. (3) presented in the Guide for Mechanistic-Empirical Design of New and Rehabilitated Pavement Structures (MEPDG), developed by National Cooperative Highway Research Program (NCHRP) [48]

$$M_r\,(MPa) = 17.6 \cdot (CBR)^{0.64} \quad (3)$$

CBR value was obtained for the colliery spoils unbound (27) and bound with cement at 2% (75). Results of CBR are showed in Fig. 10. Resilient modulus value was 145 MPa and 275 MPa for the unbound and bound with cement sections respectively. The Poisson value is chosen to take a value of 0.35 for both sections (unbound and bound), which can be a conservative value for stabilised granular materials with lime or cement [49].

For the subgrade, it assumes a resilient modulus value of 50 MPa. The Poisson ratio was of 0.35 for this layer and its thickness is undefined. According to equation (3), for this resilient modulus, value of CBR will be 5, which it is equivalent for soils types A2, A4 , A6 ,y A7 according to classification of AASHTO [50].

A summary of the characteristics materials of the designed sections types are presented in Table 14.

In the first section (unbound section), to guarantee a fully bonded between the different layers, a prime coat will be applied between the base and asphalt layer is proposed, with a dosage of 0.5 Kg/m^2, which is the minimum dosage established in PG-3 [25]. Prime coat will be composed with a cationic bituminous emulsion C50BF4, following the specifications of the UNE-EN 13808. In the second section (bound section), this same prime coat will be applied. Furthermore, a watering curing between the subbase and the granular base is proposed to avoid the rapid evaporation of the colliery spoils treated with cement and to ensure the union between these layers. A cationic bituminous emulsion C60B3 (UNE-EN 13808) with a dosage of 0.3 kg/m^2 would satisfy the regulations [25].

In the first section (Fig. 13) the unitary vertical deformation of the subbase in the upper face (εz) was of 8.64 e-4 and 5.28 e-4, under the load and between the wheels loads, respectively. According to Eq. (2), the unit deformation must be less than 9.92e-4, so the section satisfies that criterion.

In the second section (Fig 14), designed with a layer of colliery spoils bonded with cement (subbase), the maximum tensile stress (σr) in cement-treated materials and the unitary vertical deformation in the upper face (εz) in the subbase must be checked. The maximum tensile stress (σr) in the subbase was of 0.125 MPa and 0.135 MPa under the load and between the wheels loads, respectively. According to Eq (1), the maximum tensile stress must be less than 0.496 MPa, so this section meets this criterion. The unitary vertical deformation in the upper face in the subbase was of 7.15 e-4 and 3.86 e-4, under the load and between the wheels loads, respectively; these values meet the criteria of Eq. (2).

Taking into account the colliery spoils accumulated in the San José mine and the thicknesses calculated in the subbase layers (see Fig. 13 and Fig. 14), the length of roads that could be built would be 77 km and 93 km for unbound material and bound with cement respectively. If we consider what was affirmed by Skarzynska to extrapolate the results obtained to other colliery spoils in the area [9], with the existing amount of colliery spoils in the Guadiato Valley (Córdoba, Spain) we could build 328 km or 630 km for the unbound and bound sections, respectively.

4- Conclusions

The present work evaluates the use of colliery spoils collected from the *San José* mine, located in the Guadiato Valley (Córdoba, Spain), as a material for low traffic intensity road construction, both as unbound material, as bound with lime or cement. The main conclusions are detailed below:

1. Organic matter is the main limiting property of colliery spoils. The use of potassium permanganate is not an appropriate method to determine the organic matter content since this test does not take into account the mineral coal content. The use of calcination loss at 550 °C is proposed as the most appropriate method.
2. The colliery spoils are mainly composed of quartz plus some silicates and coal.
3. The colliery spoils are classified as inert according to directive 2003/33 /EC, so there is no risk of environmental contamination by the leaching of heavy metals. The high organic matter content of colliery spoils will not be a hazard to the environment.
4. Colliery spoils have certain plasticity, so it is not recommended to use them as an unbound material in the base structural layers of roads.
5. It is not recommended to use the colliery spoils as a cement-treated soil (soil-cement). A high amount of cement is necessary to achieve the minimum strength required in a soil-cement base layer.
6. The colliery spoils can be used as a subbase layer for roads, both bound and unbound. When used as a bound material with cement, the colliery spoils have a higher bearing capacity.
7. The use of colliery spoils bound with lime is not recommended, since the mechanical strength or bearing capacity does not improve.
8. Two section types of traffic category T42 (0–24 heavy vehicles per day) with unbound colliery spoils and colliery spoils bound with cement as the subbase layer have been calculated. The mixture with 2% of cement (stabilised soil with cement) allows a reduction of the thickness of this layer from 30 to 25 cm.

The results of this work will eliminate 4.2 million tons of colliery spoils collected in the old mining region of Guadiato Valley (Córdoba, Spain) and give a second life to this waste in the construction of low-traffic roads, promoting in this way the new paradigm of circular economy applied to the construction and mining sector.

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Table 1. Physico-mechanical characterization procedures

Test	Standard
Particle size distribution	UNE EN 933-1:2012 and UNE 103101:1995
Index slabs	UNE EN 933-3:2012
Dry sample density	UNE EN 1097-6:2014
Water absorption	UNE EN 1097-6:2014
Liquid Limit	UNE EN 103103:1994
Plastic Limit	UNE EN 103104:1993
Modified Proctor	UNE EN 103501:1994
Clean coefficient	UNE EN 146130:2000
Resistance to fragmentation (Angeles test)	UNE EN 1097-2:2010
California Bearing Ratio (CBR)	UNE EN 103502:1995
Particle size distribution after Proctor	UNE 103101:1995 and UNE 503501:1994
Composition of the material	UNE EN 933-11:2009
Test for free swelling of soils	UNE 103601:1996
Crushed and broken surfaces	UNE EN 933-5:1999

Table 2. Chemical characterization procedures

Test	Standard
Determination of organic matter	UNE 103204:1993
Determination of loss of ignition at 550 °C	UNE EN 15169:2007
Determination of loss of ignition at 950 °C	UNE EN 1744-1:2010
Water soluble salts	UNE 103205:2006
Gypsum content	UNE 103206:2006
Determination of total sulphur compounds	UNE EN 1744-1:2010
Determination of acid-soluble sulphates	UNE EN 1744-1:2010
Determination of water-soluble sulphates	UNE EN 1744-1:2010

Table 3. Acceptance criteria (WAC;EU Council Decision 2003/33/EC)

Parameter	Leached concentrations (mg/kg) depending of landfill class					
	Inert		**Non-hazardous**		**Hazardous**	
	L/S=2	L/S=10	L/S=2	L/S=10	L/S=2	L/S=10
Cr	0.2	0.5	4	10	25	70
Ni	0.2	0.4	5	10	20	40
Cu	0.9	2	25	50	50	100
Zn	2	4	25	50	90	100
As	0.1	0.5	0.4	2	6	25
Se	0.06	0.1	0.3	0.5	4	7
Mo	0.3	0.5	5	10	20	30
Cd	0.03	0.04	0.6	1	3	5
Sb	0.02	0.06	0.2	0.7	2	5
Ba	7	20	30	100	100	300
Hg	0.003	0.01	0.05	0.2	0.5	2
Pb	0.2	0.5	5	10	25	50
TOC	30000 mg/kg		5%		6%	

Table 4. Test of cement-bound and lime-bound with colliery spoils

Test	**Standard**	**Age**
Manufacture of test specimens	UNE 13286-51:2006	7 days
Workability	UNE-EN 13286-45:2004 and UNE 41240:2003	-
CBR index with cement	UNE 103502:1995	3 wet chamber days 4 days immerse in water
Simple compression strength	UNE 13286-41:2003	3, 7, 28, 90 days
Tensile splitting strength	UNE-EN 12390-6:2001	3, 7, 28, 90 days

Table 5. Density and water absorption

	0.063/4 mm	**4/31.5 mm**
Apparent density of particles (g/cm^3)	2.77	2.79
Particle density after drying in an oven (g/cm^3)	2.57	2.49
Density of saturated particles with a dry surface (g/cm^3)	2.61	2.61
Absorption of water (%)	4.34	3.59

Table 6. Chemical properties

Characteristic	**Value**
Organic matter. Reaction with permanganate (%)	1.92
Organic matter. Loss on ignition at 550 °C (%)	8.38
Organic matter. Loss on ignition at 950 °C (%)	9.30
Water-soluble salts (%)	0.149
Gypsum content (%)	<0.01
Total sulphur compounds (%SO_3 and %S)	<0.01
Acid-soluble sulphates (%SO_3)	<0.01
Water-soluble sulphates (%SO_3)	<0.01

Table 7. Characterisation of colliery spoils (EDX)

Characteristic	**Value (% weight)**
C (%)	9.94
O (%)	47.44
Na (%)	0.64
Mg (%)	0.76
A (%)	11.92
Si (%)	20.25
K (%)	3.37
Ti (%)	0.50
Fe (%)	5.05

Table 8. Leached concentrations (mg/kg) in material used in EURR

Parameter	**L/S= 2 l/kg**	**L/S=10 l/kg**	**Classification**
Cr	0.028	0.007	INERT
Ni	0.005	0.003	INERT
Cu	0.003	0	INERT
Zn	0.026	0.046	INERT
As	0.002	0.001	INERT
Se	0	0.003	INERT
Mo	0.005	0	INERT
Cd	0	0	INERT
Sb	0.005	0.009	INERT
Ba	0.189	0.178	INERT
Hg	0	0	INERT
Pb	0.002	0	INERT

Table 9. Summary specifications Art. 330 PG-3 and comparison unbounded use

Test	Selected Soils (ART.330.3.3.1)	Appropriate Soils (ART.330.3.3.2)	Tolerable Soils (ART.330.3.3.3)	Results	Classification
Granulometric (UNE 103101)	(# 20 > 70%) or (# 0.08 ≥ 35%)	(# 20 > 70%) or (# 0.08 ≥ 35%)	(# 20 > 70%) or (# 0.08 ≥ 35%)	#20 =89.15%	SELECTED SOILS
	D_{max}<100 mm	D_{max}<100 mm	-	D_{max}=20 mm	SELECTED SOILS
	(#0.40≤ 15 %) or (#2 <80%) (#0.4<75%) (#0.08<25%)	(#2<80%) and (#0.08<35%)		#2=21.85% #0.5<8.65% #0.125<6.1%	SELECTED SOILS
Plasticity (UNE 103103) (UNE 103104)	LL<30 and PI<10	LL<40 or (If LL>30 el PI>4)	LL<65 or (Si LL>40 the [PI>0.73(LL-20))	LL=26 PI=7	SELECTED SOILS
Determination of organic matter (UNE 103204)	OM<0.2%	OM<1%	OM<2%	1.92 %	TOLERABLE SOILS
Content in water soluble salts, included gypsum (UNE 103205)	SS<0.2%	SS<0.2%	SS (not included gypsum) <1%	0.149 % (included gypsum)	SELECTED SOILS
Test of free of swelling of soils after 4 day's soaking (UNE 103601)			Inflation<3%	0.85% (CBR)	SELECTED SOILS

Table 10. Summary specifications Art.510 PG-3 for use as well-graded crushed rock (WGCR)

Test	PG-3 Art. 510	Results	Comply
Particle size distribution (UNE-EN 933-1)	---	---	WGCR-25 and WGCR-20
Plasticity (UNE 103103) (UNE 103104)	Not Plastic	PI=7	Fails
Fragmentation resistance (Angeles coefficient) (UNE EN 1097-2)	Heavy Traffic Category T3, T4[(1)] <35	30	Satisfy
Clean coefficient (UNE 146130)	<2%	1.945	Satisfy
Form coefficient (UNE EN 933-5)	FI<35	30	Satisfy
Crushed particles (UNE-EN 933-5)	For T3 and T4[(1)] % Crushed particles	56%	Satisfy
Determination of total sulphur compounds	<1% <0.05% (layers in Contact)	0.05 %	Satisfy

[(1)] Heavy traffic category T3: 50-200 heavy vehicles/day; T4: 25-50 heavy vehicles/day, defined in Spanish Instruction of Highway

Table 11. Summary specifications Art.510 PG-3 for use as well-graded aggregate (WGG)

Test	PG-3 Art. 510	Results	Comply
Granulometric (UNE-EN 933-1)	---	---	WGCR-25 and WGCR-20
Plasticity (UNE 103103) (UNE 103104)	PI<6 Y LL<25	PI=7 and LL=26	Fails
Fragmentation resistance (Angeles coefficient) (UNE EN 1097-2)	Heavy Traffic Category T3, T4[(1)] <40	30	Satisfy

Table 12. Use as soil-cement (SC).

Test	Limit set by PG-3 Art 513	Results	Comply
Particle size distribution (UNE-EN 933-1)	SC-20 y/o SC-40	----	SC-40
Simple Compression Strength (UNE-EN-13286-51)	2.5-4.5	2.47 MPa	Fails
Workability (UNE-41240)	Full width>180 min By stripes>240 min	>240	Satisfy
Plasticity (UNE-103103) and (UNE-103104)	LL<30 PI<12	LL=26 PI=7	Satisfy
Crushed Particles (UNE-EN 933-5)	**Roads:** T00 a T1 >75 T2>50 T3 and T4 >30 **Hard shoulder:** T00 and T1 >50 T2, T3 and T4 >30	56	Satisfy For T2 and T4
Form coefficient (UNE-EN 933-3)	**Roads**: T00 a T2<35 T3 and T4 <35 **Hard shoulder**:<40	30	Satisfy For T00 and T4
Fragmentation resistance (Angeles coefficient) (UNE-EN 1097-2)	**Roads:** T00 a T2 <35 T3 and T4 <35 **Hard shoulder**:<40	30	Satisfy For T00 and T4
Organic matter (UNE 103-204)	<1%	1.92 %	Fails
Acid-soluble sulphates (UNE-EN 1744-1)	<0.8%	< 0.01 %	Satisfy

Table 13. Use as stabilized soils of colliery spoils with lime or cement

Test	Limit set by PG-3 Art 512	Results	Comply
Particle size distribution (UNE-EN 933-1)	For lime (# 80 = 100%) and (# 0.063 ≥ 15%)	# 0.063=6.5	Fails for all the types of stabilized soil with lime
Granulometric (UNE-EN 933-1)	For cement (# 80 = 100%) and (# 2 > 20%) And (# 0.063 < 35%)	# 2=21.85 # 0.063=6.5	Satisfy for all the types of stabilized soil with cement S-EST1; S-EST2 and S-EST3
Determination of Organic matter (UNE 103204)	(<2%=S-EST1) (<1%=S-EST2 and S-EST3)	1.92%	Satisfy only for S-EST1
Content in water-soluble salts, including gypsum (UNE 103205)	(<0.7%) for S-EST1, S-EST2, S-EST3	0.15 %	Satisfy for S-EST1 ; S-EST2 and S-EST3
Plasticity (UNE 103103) (UNE 103104)	PI ≤15 for S-EST1, S-EST2, S-EST3 All with cement	PI=7	Satisfy for S-EST1 ; S-EST2 and S-EST3
CBR (UNE 103502)	≥ 6 for S-EST1 ≥ 12 for S-EST1	27.4 %	Satisfy for S-EST1 ; S-EST2 and S-EST3
Simple Compression Resistance (UNE-EN-13286-51)	without limit	2.47 MPa	-
Organic matter (UNE 103-204)	<1%	1.92 %	Fails
Conglomerate content	≥ 2 % for S-EST1 ≥ 2 % for S-EST2 and S-EST3	2 %	Satisfy for S-EST1

Table 14. Summary characteristics of the proposed sections

		M_R(MPa)	Thickness (cm)	Poisson Ratio
Unbound Section (Subbase)	Pavement	1500	4	0.35
	Base	375	15	0.35
	Subbase	145	30	0.35
	Subgrade	50	Undefined	0.35
Bound Section (Subbase) 2% Cement	Pavement	1500	4	0.35
	Base	375	15	0.35
	Subbase	275	25	0.35
	Subgrade	50	Undefined	0.35

Figure
Click here to download Figure: Figures.docx

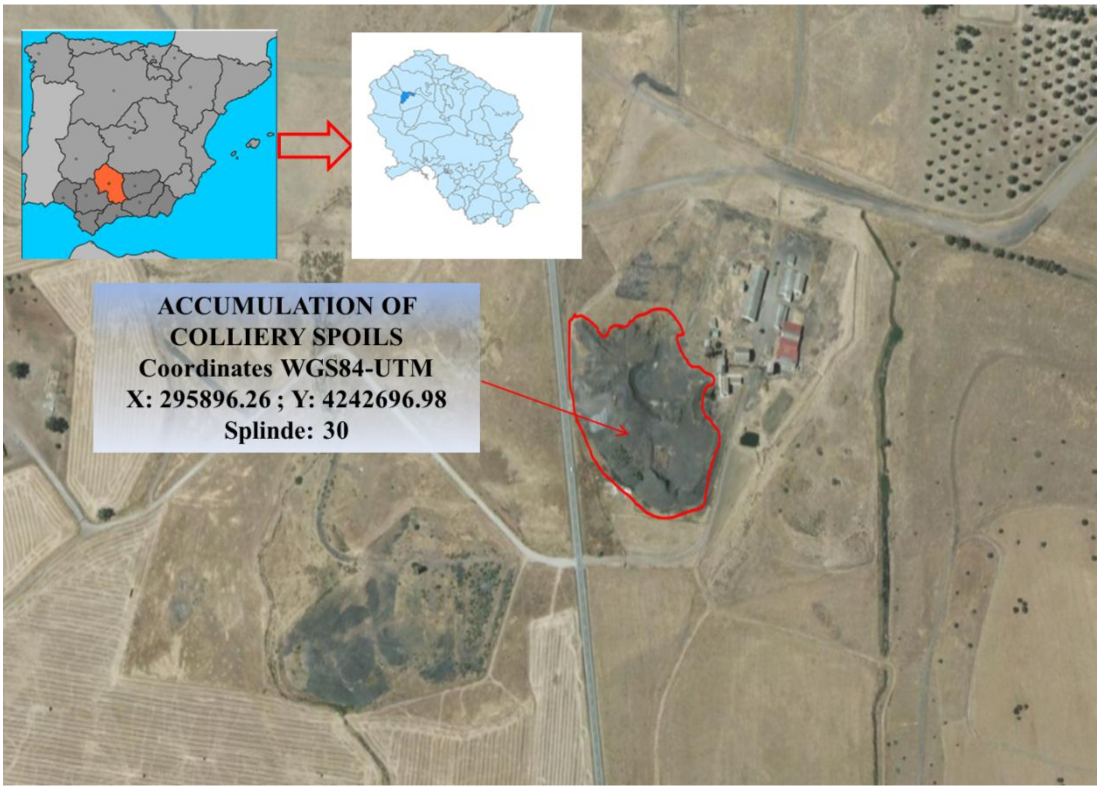

Fig. 1. Geographic situation and aerial photography of the accumulation of colliery spoils in Pozo San José (Córdoba, Spain).

Fig. 2. Stockpiling of colliery spoils in Pozo San José (Córdoba, Spain)

Fig. 3. Composition of colliery spoils**.**

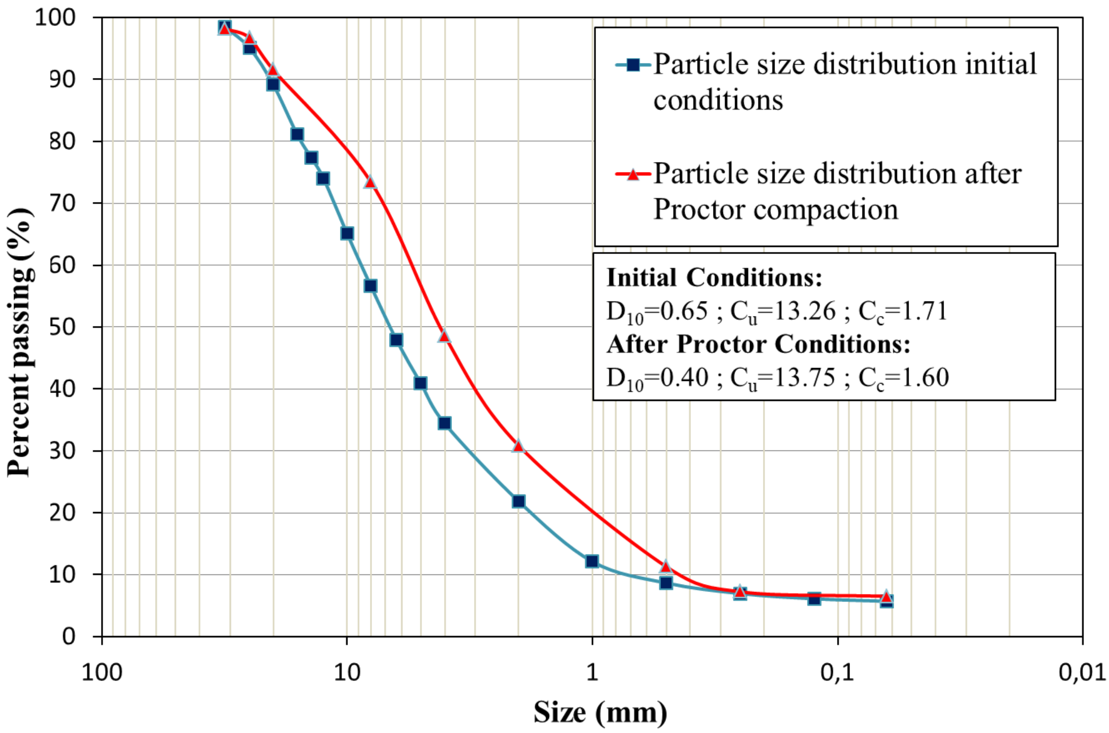

Fig. 4. Particle size distribution curves of colliery spoils before and after compaction.

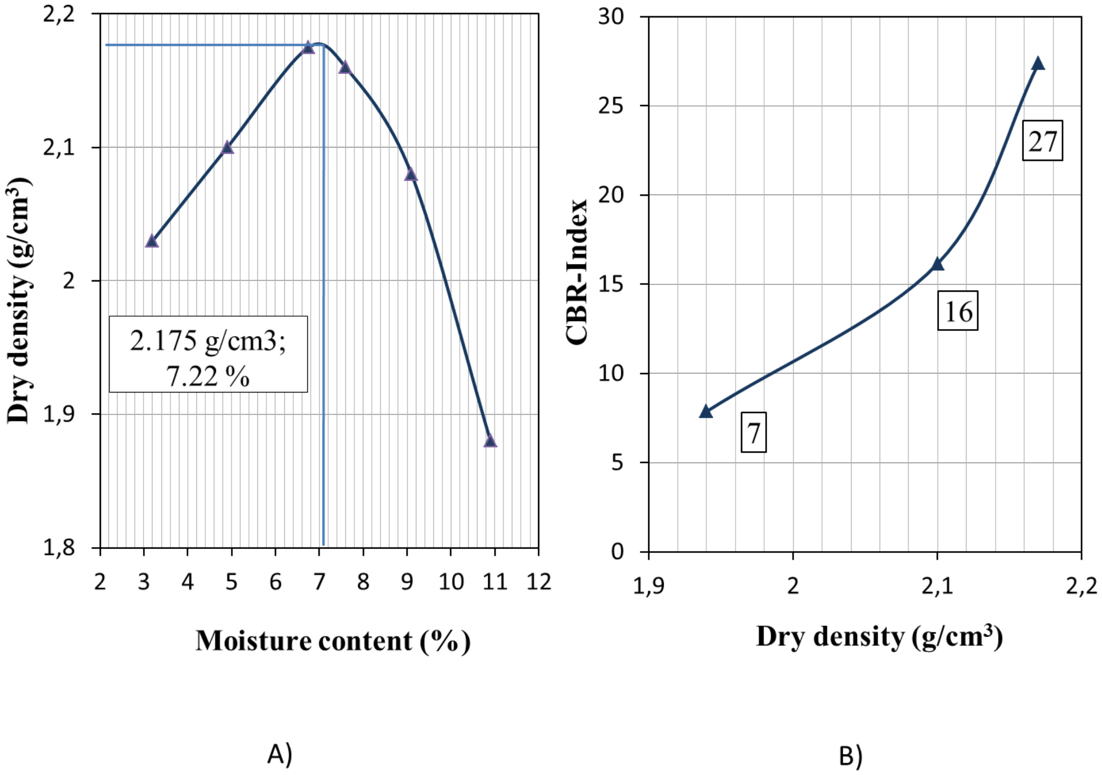

Fig. 5. A) Results of modified proctor test. B) Results of California Bearing Ratio for different dry density.

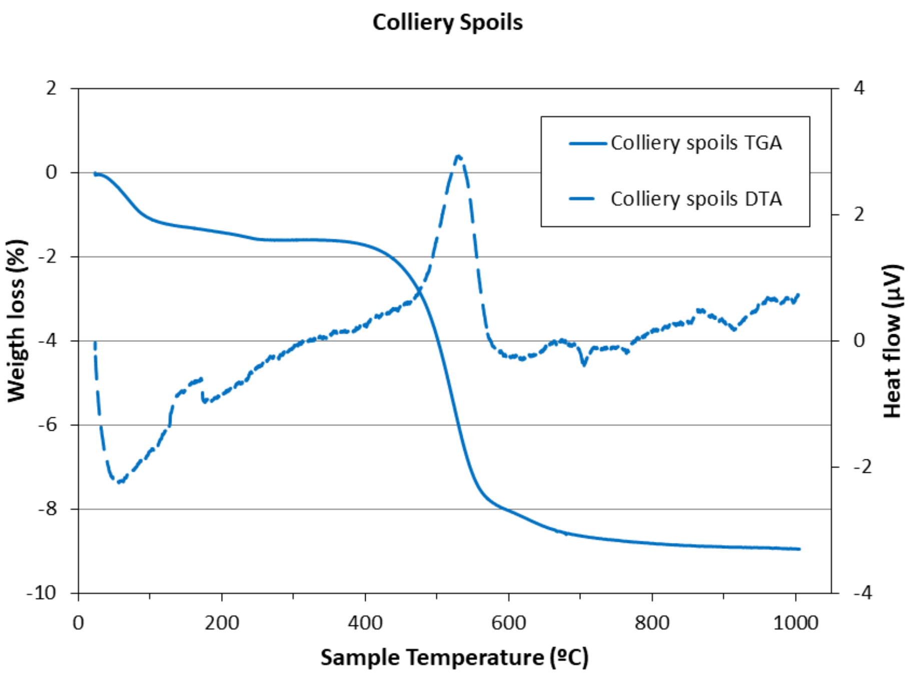

Fig. 6. TGA (dotted line) and DTA (solid lines) curves for colliery spoils.

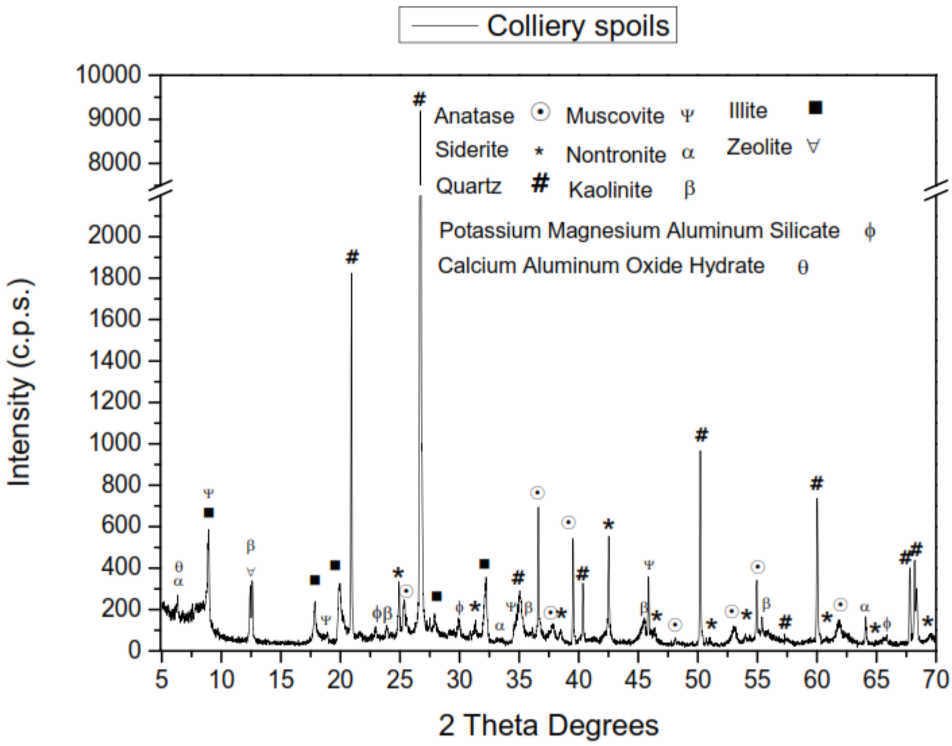

Fig. 7. XRD patterns for the colliery spoils.

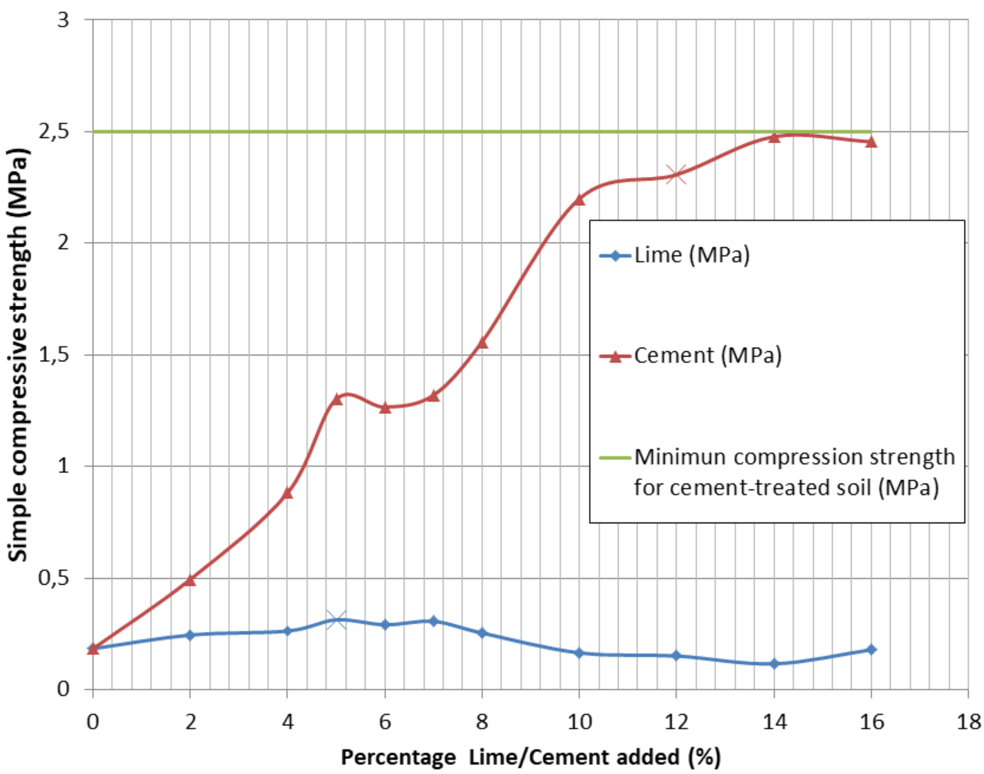

Fig. 8. Simple compression strength colliery spoils bound with cement or lime.

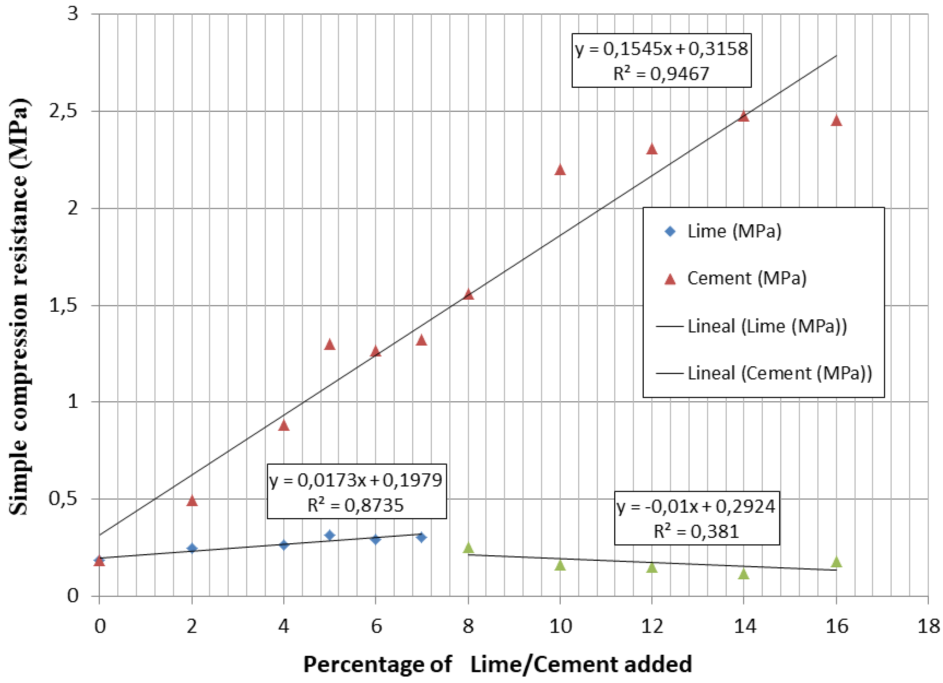

Fig. 9. Simple compression strength of colliery spoils bound with cement or lime, with trend lines.

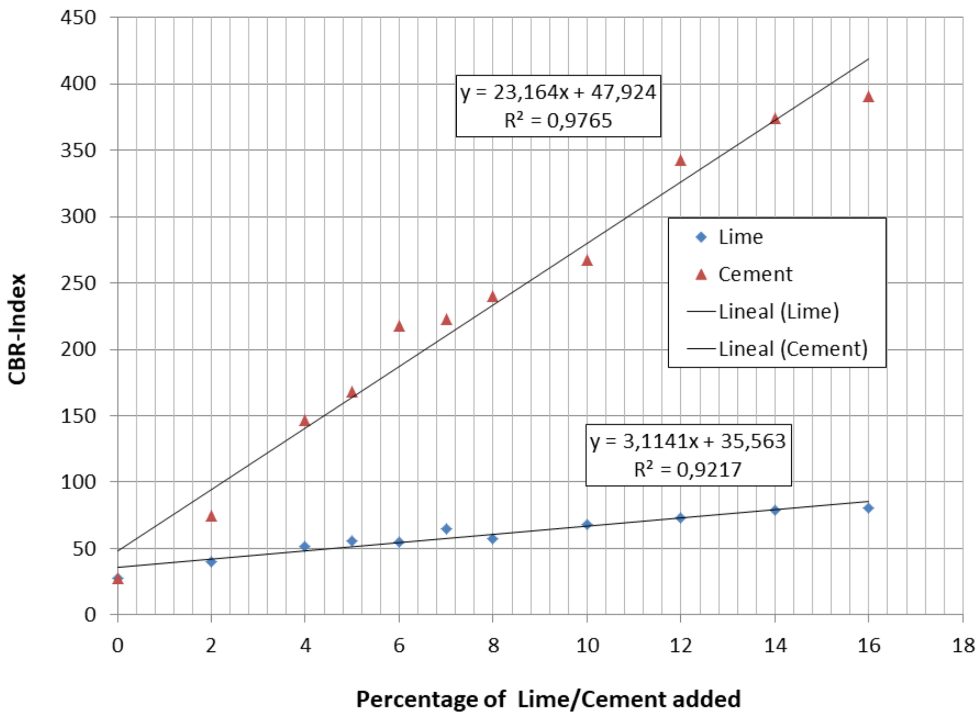

Fig. 10. Evolution of the CBR of colliery spoils bound with lime or cement.

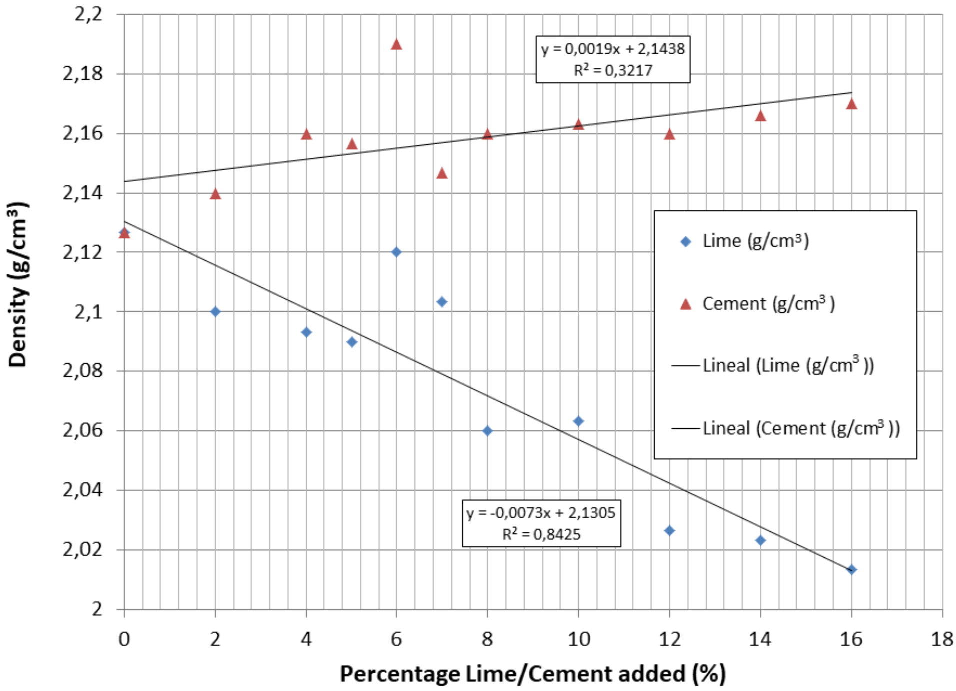

Fig. 11. Evolution of the density of colliery spoils with lime or cement.

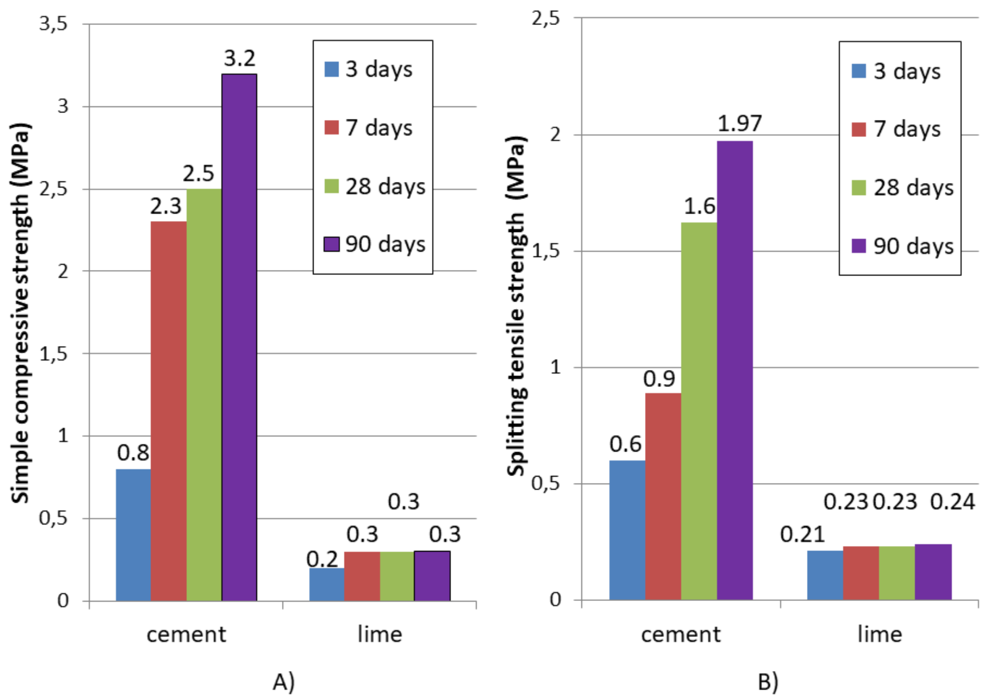

Fig. 12. A) Evolution of the simple compression strength with cement or lime at 3, 7, 28 and 90 days. B) Evolution of the tensile splitting strength with cement or lime at 3, 7, 28 and 90 days.

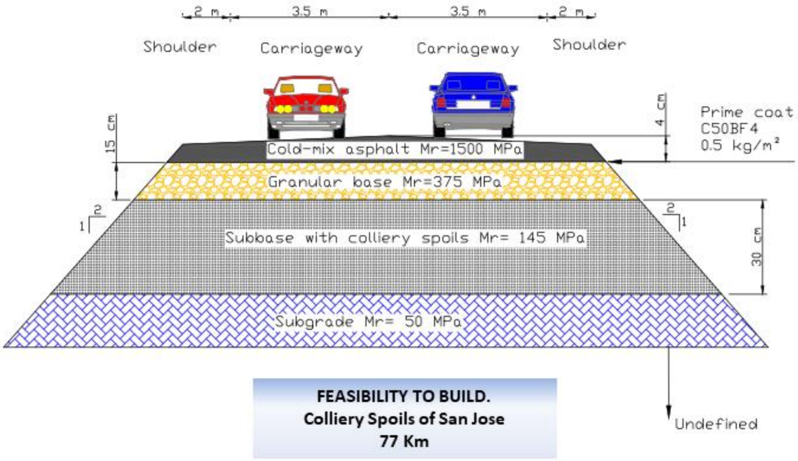

Fig. 13. Cross section of the road using unbound colliery spoils.

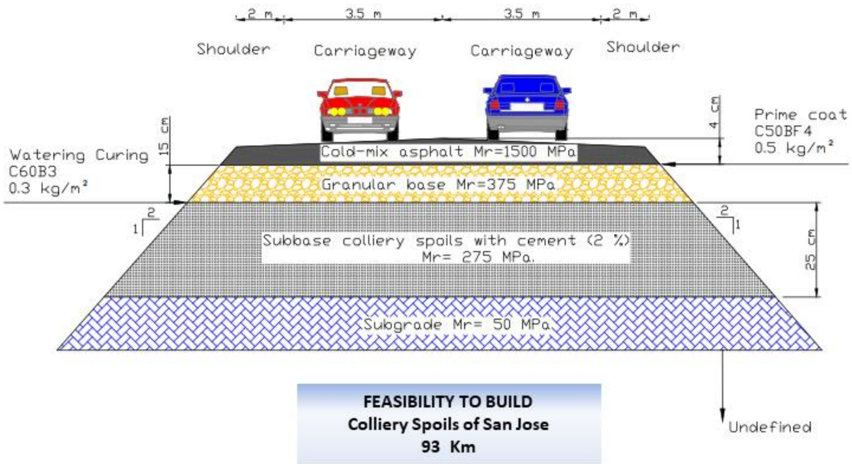

Fig. 14. Cross-section of the road using colliery spoils bound with cement.

Highlights

- Colliery spoils have been characterised for its use in road construction
- Organic matter is the main limiting property of this type of mining waste
- The use of colliery spoils mixed with lime should be discarded
- Colliery spoils can be used as a subbase layer for roads, both bound and unbound with 2% of cement
- Two section types of traffic category T42 (0–24 heavy vehicles / day) are designed