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Study on the Mechanical Behavior and Environmental Product Declarations of Road Base Layers Incorporating Anhydrous Calcium Sulphate

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Abstract. The adoption of circular economy principles and environmentally sustainable engineering is becoming increasingly prominent in civil engineering. Portland cement (PC) production alone is responsible for 8% of total annual CO_2 emissions. Novel materials should possess good mechanical and physical properties while reducing CO_2 emissions. This study analysed different mix designs for cement bound granular material (CBGM) road layers, partially replacing PC with anhydrous calcium sulphate (ACS), an industrial by-product. The objectives of the study include assessing mechanical performance through laboratory tests and analysing Environmental Product Declarations (EPDs) for each mix design. Two mix designs were examined: one with 3% ACS and 2% cement, and the other mix with 4% ACS and only 1% cement. Hydraulic binders used in the mixes included PC, Green Cement (GC), and ACS, with variations of aggregate types, natural and recycled. Innovative mix designs demonstrate comparable strengths to traditional CBGM layers. This paper aimed to analyse the environmental impacts using EPDs, comparing innovative mixes to those with 5% PC and 5% GC to identify environmentally friendly mixes with appropriate mechanical responses.

Keywords: Environmental Product Declarations, Pavement Layers, By-Product.

1. Introduction

The study aims to investigate the feasibility of partially replacing portland cement (PC) with anhydrous calcium sulphate (ACS) in cement bound granular material (CBGM) pavement layers, focusing on mechanical performance and environmental impact indicators declared in Environmental Product Declarations (EPDs). This research aligns with principles of circular economy, eco-sustainable engineering, and ecological transition, aiming to find alternatives to PC, a material known for its high energy consumption and significant CO_2 emissions during production.

Previous applications of calcium sulphate in road pavement primarily involved blends with cement, fly ash, and lime for stabilizing the soil base layer [1, 2, 3, 4, 5, 6, 7, 8]. However, this study exclusively explores the use of ACS in CBGM formulations. PC, being the most widely used construction material globally, has substantial environmental implications, with its production responsible for a considerable portion of global CO_2 emissions.

According to the International Energy Agency [9], the year 2021 witnessed the consumption of 105 kWh/t of energy for the production of 4300 Mt of cement. Cement is the construction material with the highest CO_2 emissions; its production is responsible for about 7% to 8% of the CO_2 emissions released globally. World Business Council for Sustainable Development (WBCSD) highlights how the PC preheating and calcination process generates, on average, 0.85 kg of CO_2 per 1 kg of cement [10].

In recent decades, there has been a need to reduce the negative environmental impacts of cement production; innovative methodologies were developed in raw material processing and clinker production processes, decreasing both atmospheric emissions and energy use [11, 12]. Also, the use of energy-efficient materials that can moderately replace PC in binders can be considered one of the possible methods to mitigate environmental risks [13].

In Europe, the Minimum Environmental Criteria (MEC) to be included in public tenders are treated in the National Action Plan on Green Public Procurement (PANGPP) [14]. These criteria emphasize the importance of life cycle assessment (LCA) and EPD certification in the design and use of materials in the construction works. The PANGPP outlines a framework where projects demonstrating improvements in environmental impact indicators through LCAs are incentivized in tender evaluations. This approach facilitates the greatest gains in terms of environmental sustainability, rationalizing consumption, and reducing government expenditure. LCAs and EPDs emerge as practical tools for accurate and objective measurement of the environmental impacts associated with a product or process [15].

In Italy, these tools are intended to be used following environmental policies and can serve as key parameters in tenders, as mentioned in the documents for Decree MITE 2022 [16]. In the field of road infrastructure, LCA involves detailed comparisons between materials, taking into account, for example, environmental impacts related to climate and the depletion of fossil resources [17].

By analyzing both mechanical and environmental aspects, the research aims to identify an optimal solution that balances performance requirements with reduced environmental impact.

2. Methodology

The paper presents comparative analyses of compression strength tests and EPD indicators in various mix designs to be employed in CBGM layers.

Compression strength tests were conducted after 7 days on two main mix designs: one incorporating 3% ACS and 2% PC, and the other containing 4% ACS and only 1% PC. Reference mixes, utilizing only PC as the hydraulic binder, were also analyzed for comparison. The tests were performed according to standard UNI-EN 13286-41:2021 [18]. Focusing on the most promising innovative mix, which comprised 3% ACS and 2% PC, additional compression strength tests were conducted at 28 and 75 days of curing to assess any increase in strength over time. Fig. 1 illustrates the cylindrical specimens used for conducting compression strength tests.

Fig. 1. Cylindrical specimens of CBGM incorporating ACS ready for testing.

For the EPD indicators, the innovative mix designs were compared with solutions containing 5% PC and green cement (GC) solutions. In each mix, natural aggregates (NA) and recycled aggregates (RA) were used separately.

GC is an innovative type of cement that employs fewer non-renewable resources and emits less CO_2 during its production process.

In the EPD studies, 12 mix designs were evaluated, incorporating traditional and innovative materials like PC, low carbon emissions cement, GC, ACS, NA, and RA. The environmental implications of the composite product were assessed by examining 10 impact indicators outlined in the 2019 EN 15804 standard [19]. The EPDs used are in accordance with ISO 14025 standard [20] , which is internationally recognized and developed within the International Organization for Standardization. Each EPD was provided by the respective material manufacturer: for PC, by the European Cement Association Cembureau [21]; for GC, by Italcementi [22], a member of HeidelbergCement group; and for ACS, by Fluorsid [23], a world leader in fluorochemicals for the aluminum industry.

ACS, also known as chemical gypsum, is a by-product of the chemical industry derived from the production of hydrofluoric acid.

To mitigate emissions from cement production, the decision was made to formulate mixtures for road layers that partially substitute cement with ACS.

3. Case Study

ACS, chemically known by the abbreviation $CaSO_4$, is obtained as a co-product of the chemical reaction from calcium fluoride (CaF_2) and sulphuric acid (H_2SO_4) to form hydrofluoric acid gas (HF), according to the reaction shown in Eq. (1) and Fig. 2:

$$CaF_2 + H_2SO_4 \longrightarrow 2HF + CaSO_4 \tag{1}$$

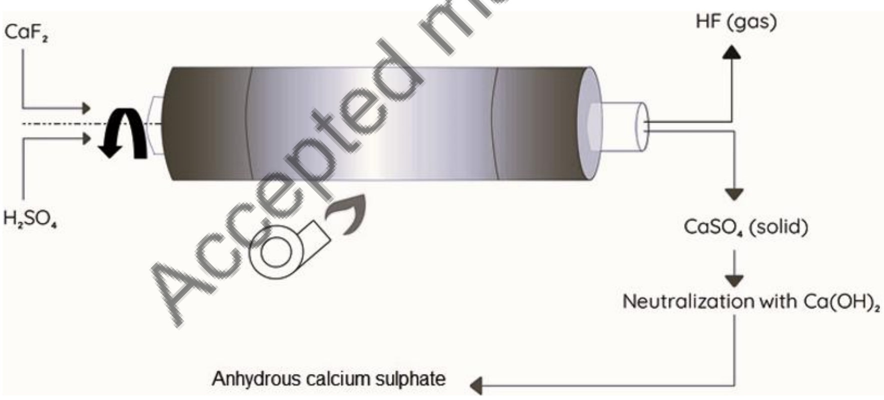

Fig. 2. Industrial production process of $CaSO_4$.

Remarkably, in this reaction, ACS constitutes 80% of the mass produced, while hydrofluoric acid makes up the remaining 20%. For every ton of hydrofluoric acid produced, there are 4 tons of ACS, resulting in a mass production ratio of 4:1 [24]. The Fluorsid industrial plant, which supplied the ACS for this study, is located on the island of Sardinia, Italy, with an average annual production of 400,000 tons. Currently, the primary commercial use of ACS is in the production of self-leveling screeds. The average market price for ACS in Italy is 50 euros per ton, while hydrofluoric acid is priced at 700 euros per ton.

3.1 Mechanical Tests

Mechanical tests were conducted in accordance with Italian specifications UNI-EN 13286-41:2021 [18], specifically compression strength tests after 7 days of curing. After 1 day in the mold, specimens were cured in wet sand with a relative humidity of 90% for 6 days. Fig. 3 presents a comparison of compressive strength tests. The initial mechanical tests were performed after 7 days, analyzing the following mixes: 3% ACS + 2% PC, 4% ACS + 1% PC, 5% ACS + 2% PC, and 3% PC.

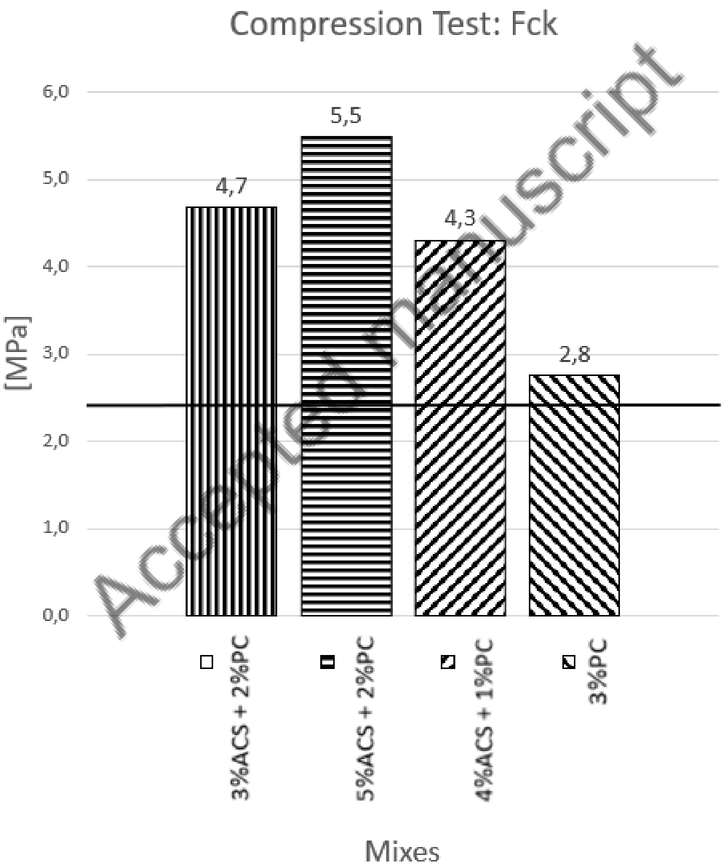

Fig. 3. Comparison of compressive strength values for various CBGM mix designs after 7 days of curing. The horizontal line marks the corresponding minimum limit of 2.5 MPa that must be achieved.

The Italian specification sets a minimum limit of 2.5 MPa for compression strength after 7 days. The highest compression strength value is achieved with the mix containing 5% ACS + 2% PC. However, this result can be attributed to the fact that this mix contains 7% binding materials, compared to the 5% and 3% found in the other mixes.

Focusing on the mechanical response over varying curing periods as depicted in Fig. 4 (7, 28, and 75 days), 32 samples of the 3% ACS + 2% PC mix were analyzed. Mechanical tests revealed that the samples' resistance improved with curing time. It was decided this research would concentrate solely on the 3% ACS + 2% PC mixes because they exhibit the most promising mechanical behavior and are also the most comparable with ordinary mixtures from a cost perspective.

Fig. 4. Comparison of cylindrical compressive strengths of 3% ACS+ 2% PC mix design after 7, 28, and 75 days of curing.

After 75 days, the compressive strength (Fck) increases by 100%. This phenomenon occurs because during the hydration process, anhydrite transforms into dihydrate gypsum, thereby enhancing its binding capacity. This is supported by Rietveld analyses that have been performed.

3.2 Environmental Impact Indicators Study

Following the mechanical testing of innovative CBGM, it is essential to analyze the environmental impact indices of the innovative solutions used. EPDs of the materials used in different mix designs were taken into account for this analysis. Each EPD was provided by the respective material manufacturer. For each mix design, the indicators of the EPDs of the various materials used were considered by mass percentage, and the impact indices associated with the quantities of material used in each mix were calculated. Twelve distinct mix designs were analyzed, each incorporating a total of 5% binder. The mix designs were categorized into two groups: ordinary mixes with 5% cement (PC and GC) [21, 22] and innovative solutions with ACS and cement in different percentages (3% ACS + 2% PC, 3% ACS + 2% GC, 4% ACS + 1% PC, 4% ACS + 1% GC). Each solution was studied using separately NA and RA [25], ensuring compliance with the specifications of standard EN 15804 2019 [19]. Ten environmental impact indices were considered: global warming potential (GWP), abiotic depletion potential of fossil resources (ADPF), soil and water acidification potential (AP), terrestrial eutrophication potential (EP-TERRESTRIAL), tropospheric ozone formation potential (POCP), stratospheric ozone depletion potential (ODP), freshwater eutrophication potential (EP-FRESHWATER), abiotic depletion potential of non-fossil resources (ADPE), seawater eutrophication potential (EP-MARINE), and water deprivation potential (WDP). Eight of these indices demonstrate that mixed designs with cement are a worse solution than those with ACS for each respective index. The worst solutions for each index are showcased in Table 1.

Table 1. Worst solutions for each index considering the three hydraulic binders used.

Index Name	GWP	ADPE	ADPF	AP	ODP	EP FW	EP TER	EP MAR	POCP	WDP
PC Mixes	o		o	o			o		o	
GC Mixes					o	o				o
ACS Mixes		o						o		

Global warming potential (GWP), abiotic depletion potential of non-fossil resources (ADPE), abiotic depletion potential of fossil resources (ADPF), soil and water acidification potential (AP), stratospheric ozone depletion potential (ODP), freshwater eutrophication potential (EP-FW), terrestrial eutrophication potential (EP-TER), seawater eutrophication potential (EP-MAR), tropospheric ozone formation potential (POCP), water deprivation potential (WDP).

As depicted in Table 1, ACS solutions perform poorly only in terms of ADPE and EP MAR indices. Table 2 reports mix designs categorized into two families based on whether they utilized NA or RA. The labels shown in the subsequent graphs are given accordingly and correspond to those in Table 2.

Table 2. Label specification used for each of the analysed mix designs.

Mix Design	5 PC	5 GC	3 ACS + 2 PC	4 ACS + 1 PC	3 ACS + 2 GC	4 ACS + 1 GC
Natural Aggregates (NA)	5% Portland Cement	5% Green Cement	3% Sulphate + 2% Portland Cement	4% Sulphate + 1% Portland Cement	3% Sulphate + 2% Green Cement	4% Sulphate + 1% Green Cement
Recycled Aggregates (RA)	5% Portland Cement	5% Green Cement	3% Sulphate + 2% Portland Cement	4% Sulphate + 1% Portland Cement	3% Sulphate + 2% Green Cement	4% Sulphate + 1% Green Cement

Analyzing the 10 environmental impact indices for all 12 mixes revealed that, in 80% of cases, cement solutions are not recommended compared to ACS solutions. As illustrated by the following graphs, the studied indices include GWP reported in Fig. 5, ADPF reported in Fig. 6, POCP reported in Fig. 7, and EP FW reported in Fig. 8, which are particularly significant for the analysis.

Fig. 5 illustrates how traditional PC solutions are the most impactful. Conversely, the optimal solution is 4 ACS + 1 GC and RA, which has a lower impact. This is because ACS is less impactful than GC, and GC is less impactful than PC. In terms of this index, there is no significant difference when using NA or RA in the mix.

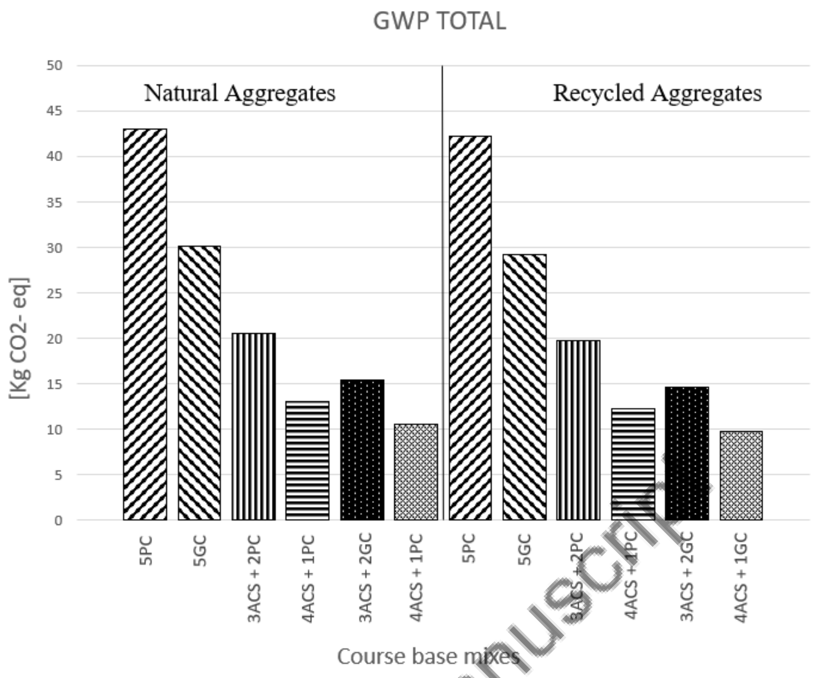

Fig. 5. Comparison of the mix designs impacts according to GWP Total Index.

Similarly, in Fig. 6, the mix designs using PC alone are the most impactful (5 PC with NA and 5 PC with RA), while options with ACS + GC are significantly less impactful. In fact, for this index, ACS is more environmentally sustainable than GC. For this reason, the least impactful solution is 4 ACS + 1 GC with RA because a great amount of ACS is used.

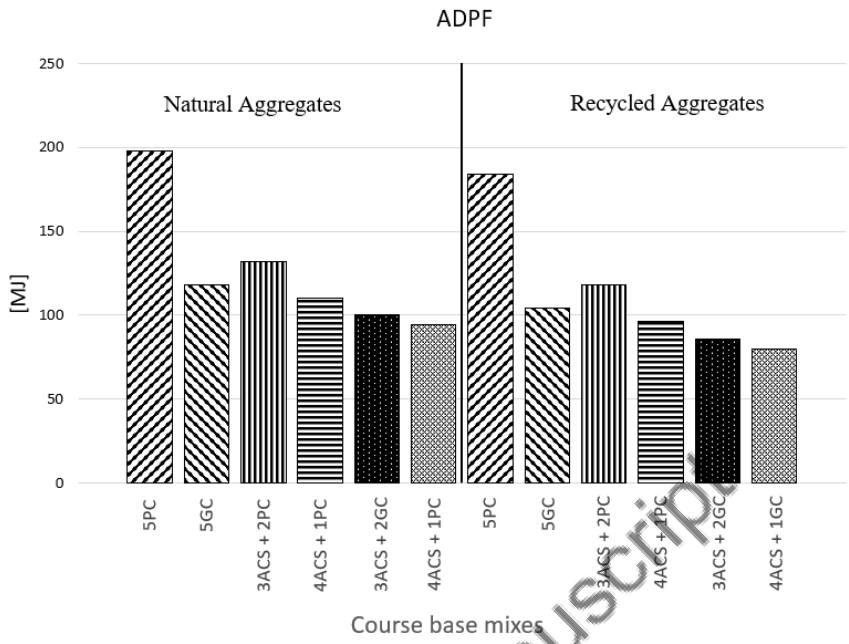

Fig. 6. Comparison of the mix designs impacts according to ADPF Index.

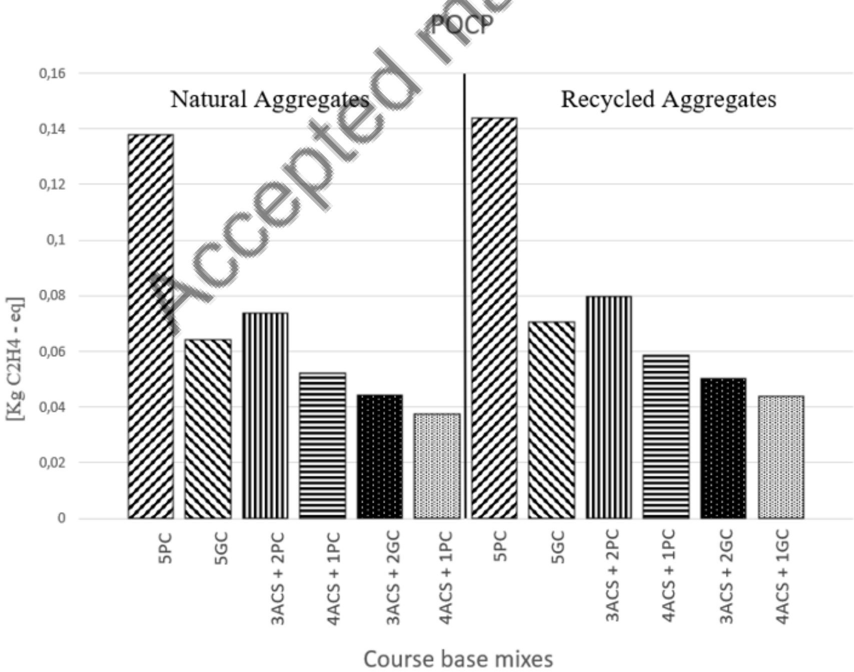

Fig. 7. Comparison of the mix designs impacts according to POCP Index.

As depicted in Fig. 7, solutions demonstrating the highest impact involve the use of PC (5 PC with NA and 5 PC with RA). Conversely, the lowest impacts are observed when GC and ACS are employed, particularly in cases where the ratio is 4% and 1% (4 ACS + 1 GC with NA, 4 ACS + 1 GC with RA). This outcome may be attributed to the lower impact of GC compared to PC and the favorable environmental characteristics of ACS.

In Fig. 8, the most impactful solutions are those in which GC is used (5 GC with NA and 5 GC with RA). ACS has a higher EP FW index than PC but lower than GC. For this index, the least impactful solution is that using only PC. In fact, it is possible to see how the innovative solutions are not eco-sustainable. In this case, RA has a lower impact than NA.

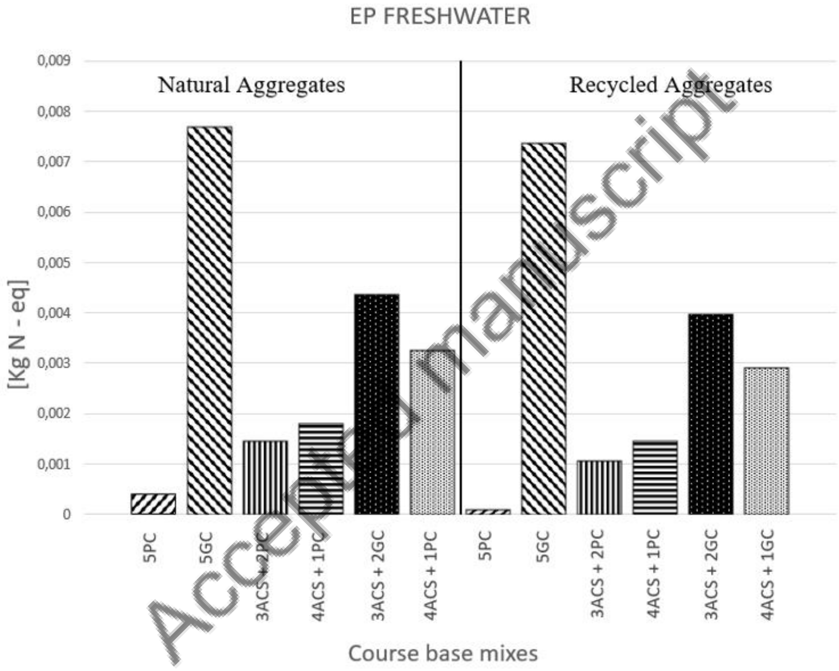

Fig. 8. Comparison of the mix designs impacts according to EP FW Index.

4. Results And Discussion

The exclusive use of ACS in the road base layers has been infrequently employed. More commonly, ACS is employed in the road pavement layer in asphalt mixes or to stabilize the soil base layer. Compression tests conducted after 7 days indicate that all the innovative mixes studied can withstand stress exceeding 4.3 MPa, in compliance with the specified requirements. The mix design incorporating 3% ACS + 2% PC demonstrates a compression strength of 4.7 MPa after 7 days. Subsequent compression tests performed over time on this mix design reveal that after

75 days, its resistance can improve by 100%. This innovative mix enhances its mechanical strength due to the ongoing anhydrite hydration reaction, resulting in dihydrate gypsum. Analyses conducted in parallel regarding the environmental impact indices of the different mix designs achievable by combining ACS, PC, and GC in different percentages with RA and NA show that 70% of the innovative solutions are preferable to those using only cement. Results are resumed and summarised in Table 3: "V" tick indicates the best eco-sustainable solution:

Table 3. Best eco-sustainable solutions for each index considering the hydraulic binders used.

Index Name	GWP	ADPE	ADPF	AP	ODP	EP FW	EP TER	EP MAR	POCP	WDP
PC Mixes		V				V				
GC Mixes								V		
ACS Mixes	V		V	V	V		V		V	V

Global warming potential (GWP), abiotic depletion potential of non-fossil resources (ADPE), abiotic depletion potential of fossil resources (ADPF), soil and water acidification potential (AP), stratospheric ozone depletion potential (ODP), freshwater eutrophication potential (EP-FW), terrestrial eutrophication potential (EP-TER), seawater eutrophication potential (EP-MAR), tropospheric ozone formation potential (POCP), water deprivation potential (WDP).

It may seem inconsistent that, as shown in Table 1, 80% of the worst solutions contain cement. In Table 3, the best solutions are highlighted, with ACS present in 70% of them. This discrepancy arises because, for the EP FW index, the contrast between cement and sulphate is not respected; instead, the best solution involves PC while the worst involves GC. In Table 4, the abbreviation "RA" signifies that the better solution is achieved using recycled aggregates, while "NA" indicates the use of natural aggregates.

Table 4. Best solutions for each index considering only innovative mix design with ACS.

Indexes	GWP	ADPF	AP	ODP	EP TER	POCP	WDP
3%ACS+2%PC				RA			RA
4%ACS+1%PC							
3%ACS+2%GC							
4%ACS+1%GC	RA	RA	NA=RA		NA	NA	

Global warming potential (GWP), abiotic depletion potential of fossil resources (ADPF), soil and water acidification potential (AP), stratospheric ozone depletion potential (ODP), terrestrial eutrophication potential (EP-TER), tropospheric ozone formation potential (POCP), water deprivation potential (WDP).

It can be observed that for most indices, the better solution is 4% ACS + 1% GC. The use of RA is slightly preferable to NA.

5. Conclusion

The application of ACS in road bases, particularly as a partial substitution for PC, demonstrates significant potential. The specification limits are fully met after 7 days. Samples with 3% ACS and 2% PC percentages show a 100% increase in compression resistance after 75 days. On the other hand, the study on Environmental Impact Indices shows that innovative solutions with ACS in CBGM are less impactful than ordinary solutions with only cement use in 70% of cases. Further studies will be performed to evaluate further mechanical performances like elastic moduli and indirect tensile strength. In addition, a leaching test will be conducted on the mixes to evaluate environmental compatibility.

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