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Article

Preparation of Green Sustainable Cement Paste Mixture Based on Inorganic Additives: An Experimental and Modelling Approach

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Abstract: Using waste materials in the mixture of building materials is an approach aligned with the circular economy, a viewpoint that creates sustainable building industries, especially in developed countries. This study concentrated on the application of laponite (LAP), fly ash (FA), and bentonite (BENT) materials in the mixture of cement pastes. The first step used experimental practices to examine the metrics of toughness, three-point bending, and compressive strength with different percentages of added LAP, FA, and BENT after the characterization of samples by scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS). The next step entailed assessment of cement paste specifications through some regressive equations obtained by the application of 2D curve fitting and sensitive analysis of additive (FA, LAP, and BENT) fluctuations in the structure of cement paste. The results show that linear polynomial equations are the best for the evaluation of cement paste terms as per different percentages of the additives. The environmental impact assessment (EIA) of nine prepared samples demonstrated that LAP created the safest condition in comparison to others. However, the ordered weighted averaging (OWA) computations applied for the sustainability assessment (SA) of the samples showed that the LAP is the most appropriate option for use in the structure of cement paste. Using experimental analysis and mathematical modeling, the behavior of cement paste interacting with mineral additives is evaluated. Sustainable mixtures are then presented based on EIA.

Keywords: sustainability; environmentally friendly cement paste; environmental impact assessment; mineral additives; sensitive analysis; ordered weighted averaging

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1. Introduction

Research on the application of green materials in cementitious composites is of vital importance in today's world. The construction industry is a significant contributor to environmental degradation, consuming vast amounts of natural resources and emitting substantial greenhouse gases [1]. Countries from all around the world are paying more attention to the disclosure of data regarding the construction and building industry's use of energy, materials, and water and waste generation, which has prompted them to give this problem top priority on their political agendas. The European Union (EU) has specifically acknowledged the compelling evidence revealing that the building sector alone accounts for 42% of the EU's total final energy consumption, over 50% of extracted materials, 30% of water consumption and waste creation, and 35% of greenhouse gas (GHG) emissions. [2]. Therefore, finding sustainable alternatives to traditional construction materials is crucial for mitigating these environmental impacts. Research in this field is important in several key aspects. First and foremost, it enables the development and implementation of more sustainable construction practices. By exploring and understanding the potential of green materials, researchers can contribute to reducing the carbon footprint associated with cementitious materials [3]. This research can lead to the discovery of innovative and eco-friendly materials that possess similar or enhanced properties compared to conventional materials. Furthermore, research on the application of green materials in cementitious materials fosters the development of a circular economy [4,5]. It promotes the use of recycled or waste materials as substitutes for conventional components, reducing the reliance on virgin resources and minimizing waste generation. This not only conserves natural resources but also decreases the environmental impact associated with the extraction and processing of raw materials [6]. Moreover, the research in this field can contribute to improved energy efficiency in the construction industry. Green materials often require lower energy inputs during manufacturing or can be produced using renewable energy sources. Investigating and optimizing the manufacturing processes of green materials can result in significant energy savings and a more sustainable construction sector [7].

Cement production is a significant contributor to carbon dioxide emissions due to the energy-intensive process of clinker production. The process of manufacturing Portland cement is known to generate carbon dioxide emissions that are nearly equivalent to each unit of cement produced. Moreover, besides the substantial carbon dioxide emissions associated with concrete production, it also requires a significant amount of materials, leading to strain on resource deposits and environmental degradation. Specifically, the production of Portland cement consumes approximately twice the volume of raw materials required to produce one ton of cement [8]. Additionally, the global cement industry produces approximately 30 million tons of a solid waste, known as cement kiln dust, annually [9]. Therefore, it is important to find alternative materials for replacing cement partially or completely, reducing the environmental impact of cementitious materials. In this context, the replacement of cement with waste materials and clays is a promising avenue of research [10]. Waste materials such as fly ash, slag, and silica fume are commonly used as cement replacements [11]. These materials are byproducts of industrial processes, such as coal combustion or metal smelting, that would otherwise be disposed of as waste. As an example, roughly 50% of coal ash is currently disposed of as waste in the United States, 7.1% in the European Union (EU), 30% in China, and 75% in India [12]. Incorporating these waste materials into cementitious mixtures not only reduces the demand for cement but also provides a solution for their disposal and helps to conserve natural resources. Some clays, such as calcined clay or metakaolin, have pozzolanic properties, meaning they react with calcium hydroxide to form additional cementitious compounds [13]. These clays can be used as partial replacements for cement, enhancing the strength and durability of the resulting cementitious materials. Moreover, clays are abundant and widely available, making them a sustainable alternative to cement [14,15].

Assessing the sustainability of green cement paste mixtures involves evaluating multiple criteria to ensure that the material contributes positively to environmental, economic, and social aspects. One primary criterion is the reduction of carbon emissions, achieved by partially replacing Portland cement with supplementary cementitious materials (SCMs) like fly ash, laponite, and bentonite. Fly ash, a byproduct of coal combustion, and other industrial waste materials reduce the need for clinker production, thereby lowering CO_2 emissions significantly [16]. Another important criterion is resource efficiency, where the use of industrial byproducts helps minimize waste and promotes the recycling of materials, thus conserving natural resources. Additionally, the durability and longevity of the green cement paste are critical for sustainability. Improved mechanical properties and resistance to environmental degradation, as evidenced by the inclusion of laponite and bentonite, enhance the service life of structures, reducing the need for frequent repairs and replacements [17]. Economic viability is also assessed through cost–benefit analysis, considering the long-term savings from reduced material costs and maintenance. Furthermore, the health and safety impacts on workers and end-users are evaluated to ensure that the materials do not pose any adverse health risks. Finally, lifecycle assessment (LCA) is used to holistically evaluate the environmental impacts of the cement paste from production to disposal, ensuring that the overall environmental footprint is minimized [18].

The use of green materials in construction and work in the building industry that involves replacing cement with more sustainable materials (like industrial wastes and clays) have the aim of reducing carbon emissions, decreasing waste generation, and improving the sustainability of cementitious materials. This not only contributes to greener construction practices but also addresses the global challenges of resource depletion and climate change.

Furthermore, the size of the green buildings market, as demonstrated in Figure 1, has increased over the last few years [19]. According to the scheme, the USA is the frontier of this field, and it will be developed in different regions of the world in the coming years. With the growth of green building industries in developed and developing countries, the environmental impacts of industrialization, such as global warming, waste emission, and health risks, can be addressed.

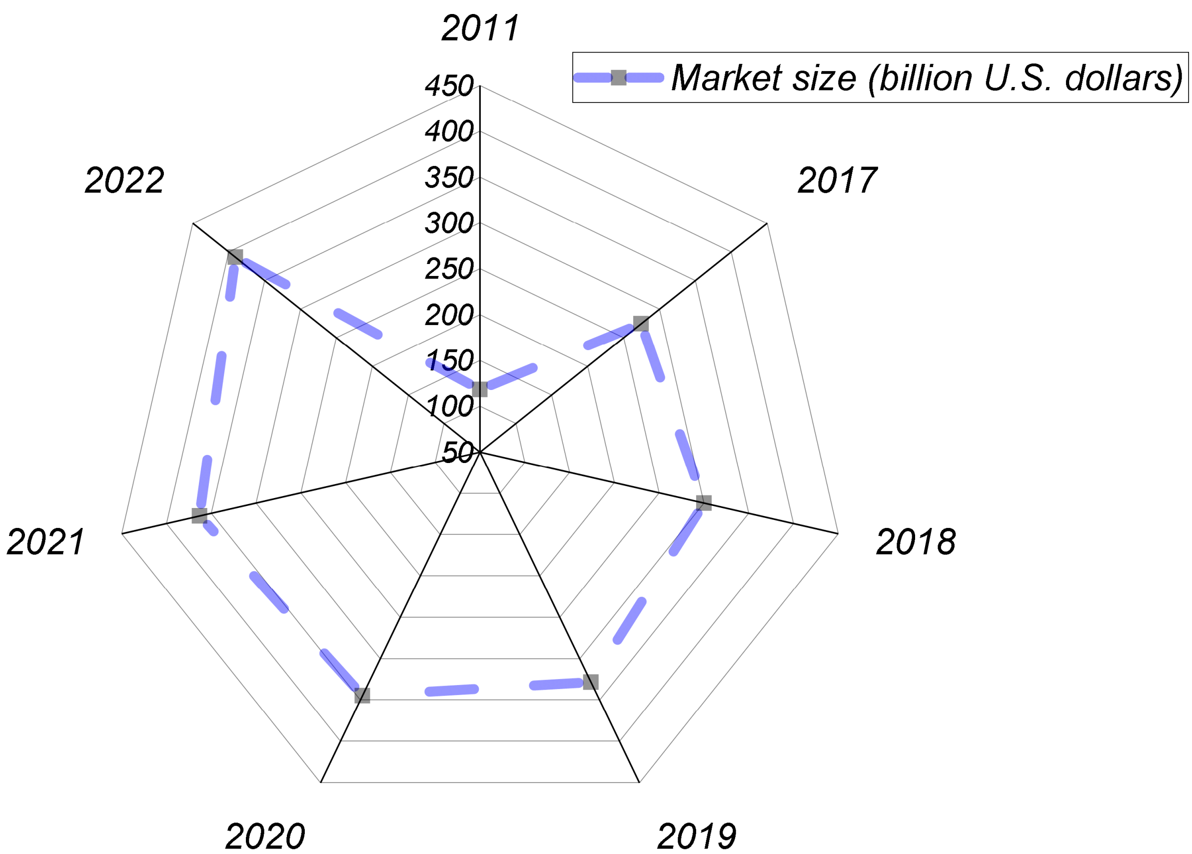


Figure 1. The statistical outcomes of green building market size fluctuations in the USA: 2011–2022.

There are lots of research items in the field of green cement preparation that have been developed from various perspectives. The most recent studies are presented in Table 1.

Table 1. Summarization of our literature review in the field of green material applications in cement industries.

Tools and Materials	Goals	Researchers
Experimental application tests and characterization of prepared concrete in different mixtures	The evaluation of blast furnace slag additive in the cement in time series	[20]
Micro and macro experiments on biochar low-carbon cement mixtures to estimate hydration and engineering properties	Assessment of biochar–limestone calcined clay in low-carbon cement at different ages	[21]
Optimization of geo-polymer concrete samples with different concentrations	Influence of ceramic waste powder and PVA on geo-polymers	[22]
Experimental design, characterization and mechanical tests for different curing temperatures and at different ages	Impact of curing temperature of alkali-activated laterite-rock-powder-based geo-polymer samples	[23]
Experimental evaluation and characterization of cement-based composites by adding different materials	Improvement of thermoelectric properties of large-sized thermoelectric cement composites for surface temperature reduction and pavement energy harvesting	[24]
Optimization of mechanical and durability properties and characterization of samples with different percentages of materials	Enhancement in mechanical properties and durability of high-strength concrete with wheat straw ash as a partial replacement for cement	[25]
Characterization of the samples by using XRD, SEM, TEM, BET and MIP	Study of mortars replaced with waste agriculture waste in the form of aerogel	[26]
Investigation of the predictive performance of various machine learning models for estimating compressive strength	Determination of compressive strength of green concrete with blast furnace slag	[27]
Mechanical tests of the samples with different dosages of rice straw ash	Evaluation of self-compacting concrete with rice straw ash as a partial replacement for cement at different time periods	[28]
Physical and mechanical testing of the samples containing different percentages of calcined sludge	Study of cementitious materials with partial replacement of cement by sludge at different ages	[29]

A novel ultrasonic treatment using graphene quantum dots (GQDs) significantly improves the dispersion and exfoliation of 2D nanomaterials (GO, CLDH, CN), enhancing their ability to accelerate cement hydration and improve mechanical properties. This method increases the specific surface area and provides more nucleation sites, leading to better cement composite performance [30]. Additionally, research into ternary cementless composites using red mud (RM), ultra-fine fly ash (RUFA), and ground granulated blast-furnace slag (GGBS) reveals that while RM increases setting time and reduces fluidity and compressive strength, GGBS enhances compressive strength and alters hydration products, with optimal performance observed in a mix containing high GGBS content, achieving a compressive strength of 47.3 MPa [31]. Durrant et al. investigated the sorption and desorption processes of cesium in binary and ternary mineral systems over long periods, revealing that while cesium sorption to montmorillonite and kaolinite is reversible, sorption to illite shows partial irreversibility, possibly due to slow desorption kinetics rather than permanent fixation [32]. Furthermore, incorporating varying amounts of reactive MgO into mortars and optimizing the curing regime with carbonation and standard curing significantly improved pore size distribution, chloride ion resistance, and mechanical properties. However, excessive MgO led to $Mg(OH)_2$ formation, causing cracks and reduced strength, thereby accelerating CO_2 diffusion and lowering the pH [33].

With concentration on Table 1, it is clear that most of the studies in the field of green cement and concrete invention are focused on characterization and physio-mechanical assessment of the materials performance. Despite these advancements, there is a gap in the comprehensive evaluation of the combined use of fly ash, laponite, and bentonite as partial replacements for cement. This study aims to address this gap by investigating the effects of these additives on the mechanical properties and durability of cementitious composites, thereby providing new insights into optimizing eco-friendly and high-performance building materials. According to Table 1, it can be understood that the application of statistical optimization computations for sensitive analysis is a rare field which has been assessed by the present research work. For support data for the evaluation of gaps in the present research, a literature review was based on VOSViewer software version 1.6.20 and the Scopus database. After searching the Scopus database, 182 research records were categorized in the field of green cement. Likewise, the 20 repetitions of the selected keywords were determined for further assessment, as demonstrated in Figure 2. Based on the scheme (Figure 2), we know that the application of fly ash as a partial replacement for cement has been considered in previous research. Moreover, the combination of fly ash, bentonite, and laponite in research is more rare; this will be specifically analyzed in this study.

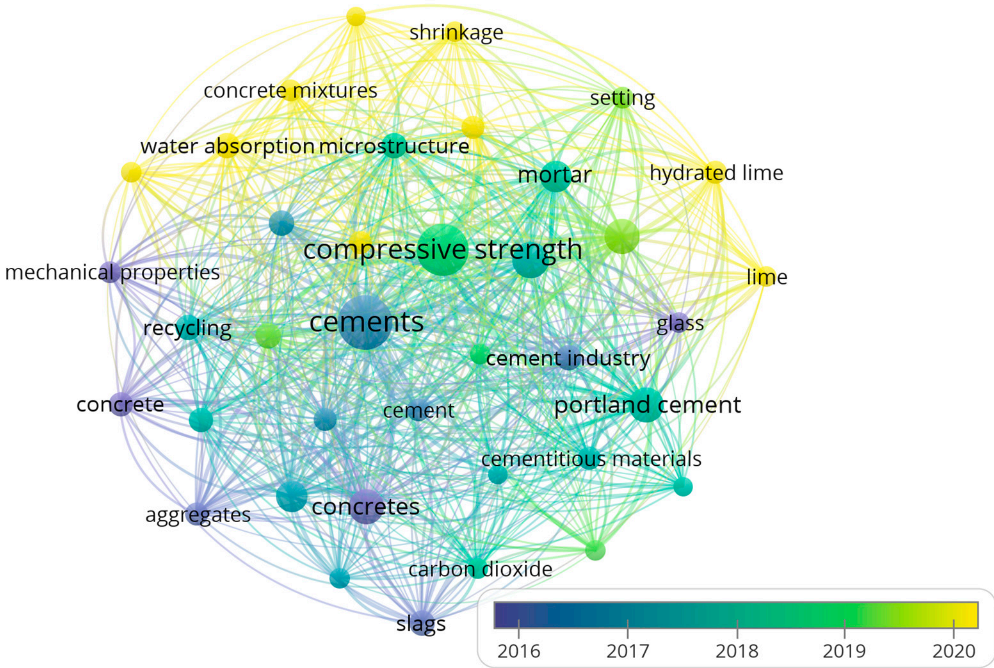


Figure 2. The schematic plan of keyword occurrence based on scientometric analysis of green cement: VOSViewer software.

This study aims to present (I) preparation of green cement paste samples with different added percentages of fly ash (FA), bentonite (BENT), and laponite (LAP); (II) a characteristic analysis of sample behaviors using some experimental instruments; (III) a statistically sensitive analysis of the most important features of three-point bending stress, toughness, and compressive strength; (IV) a statistical regression assessment of FA, BENT, and LAP correlated by all functions; an (V) environmental impact assessment based on concentrations of toxic compounds in the additive compounds; and (VI) a method for assessment of the sustainability of prepared mixtures via the application of the ordered weighted averaging (OWA) method.

In the following, all materials and methods, including applied experimental protocols, sample preparation, utilized instruments, and mathematical models, are expressed in Section 2. Likewise, Section 3 conveys the main results and arguments with other research practices. Finally, the most significant achievements of this study are presented in Section 4.

2. Materials and Methods

The research roadmap of the present research is depicted in Figure 3. According to the roadmap, in the first step, different percentages of FA, BENT, and LAP are added to different samples of cement paste, and then the performance of the samples are analyzed based on three cost functions, including three-point bending stress, toughness, and compressive stress. While the specifications of each sample are evaluated by scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS) experiments. In the next step, the obtained data are examined with the application of regression modelling due to sensitive analysis and mathematical modelling. Then, prepared samples are evaluated based on their performance, investment costs, and toxic elements utilizing OWA.

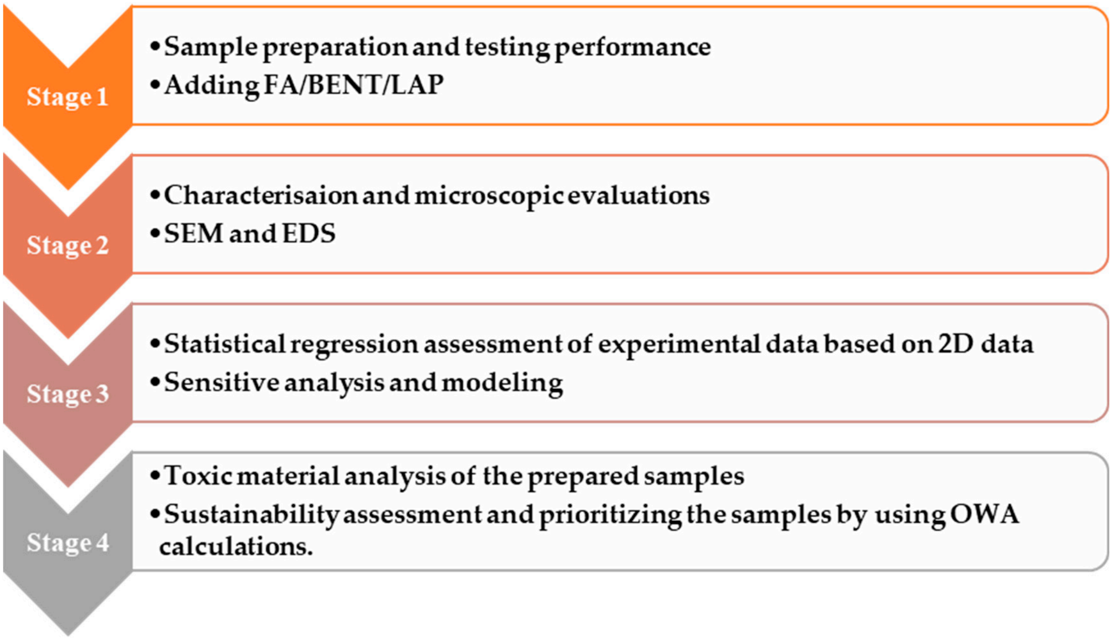


Figure 3. The research roadmap of the investigation.

2.1. Applied Materials

In the present research, all applied materials are summarized in Table 2 based on their specifications.

Table 2. The specifications of the applied materials in the study.

Material	Specification
Cement	Ordinary Portland cement ČSN EN 197-1, Denmark
Fly ash	Fly ash for concrete as per DIN EN 450, Betoment OP Germany
Laponite RD	SYnL-1 (Synthetic layered silicate, hydrous sodium lithium magnesium silicate), Clay Minerals Society Source Clays Repository P.O. Box 460130, Aurora, Colorado 80046-0130 USA
Bentonite	Bentonite Clay Ekokoza s.r.o, Czech Republic

The bulk density and surface area of fly ash range between 0.54–0.86 g/cm^3 and 300–500 m^2/kg, respectively [34]. The bulk density and surface area of laponite have been reported as 1 g/cm^3 and 370 m^2/g, respectively [35]. The density and surface area of bentonite have been reported as 2.5 g/cm^3 and 152 m^2/g, respectively [36].

The details of geometrical characteristics of particles and SEM images are discussed in Section 3.

2.2. Experimental Protocols and Samplings

Due to experimental activities in the present investigation, three protocols are applied for sample preparation using a mixer (ČSN EN 1008 (732028)) [37], determination of flexural and compressive strength of hardened mortars (ČSN EN 1015-11 (722400)) [38], and purpose of impact strength by the Charpy method (ČSN EN ISO 179-2 (640612)) [39]. Meanwhile, the stages of sample preparation and experimental practices are declared in Figures 4 and 5, respectively. According to Figure 4, in the first step, different samples were mixed with individual formulations demonstrated, as per Table 3. It should be mentioned that in all the samples in Table 3, the water to binder ratio (WBR) was equal to 0.4. Then, the prepared samples were cast with simple rectangle and cube shapes following the shaking of the samples for uniform distribution and compaction. Then, the samples were cured for 28 days in standard condition. Based on Figure 5, the applied materials were characterized by both SEM and EDS tests. Likewise, the cured samples were utilized for three mechanical tests including three-point bending, toughness, and compression tests in the lab.

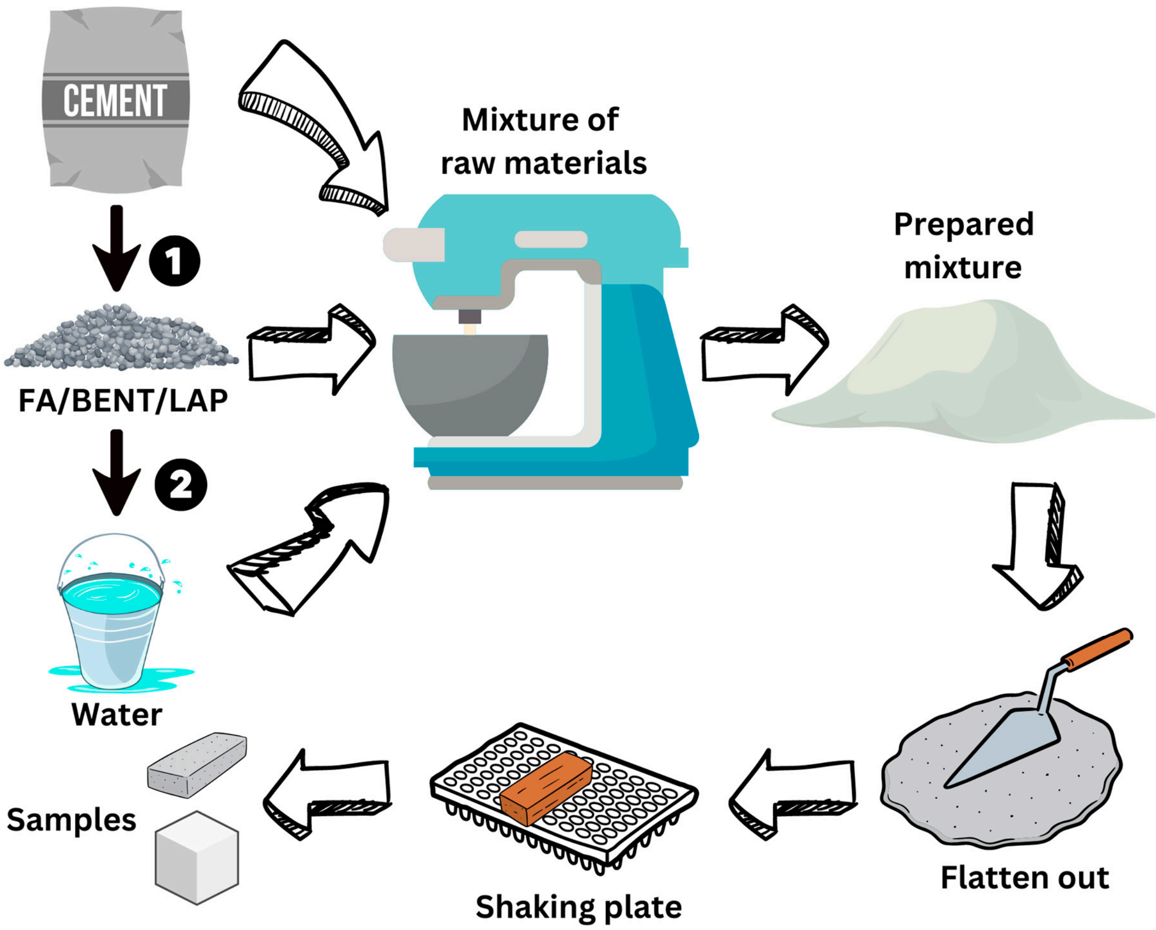


Figure 4. The process of sample preparation in this research.

Table 3. Samples with different fillers in the present study.

Sample Name	Fly Ash (FA)	Laponite (LAP)	Bentonite (BENT)
Sample 1 (S1)	5%	0	0
Sample 2 (S2)	10%	0	0
Sample 3 (S3)	20%	0	0
Sample 4 (S4)	0	1%	0
Sample 5 (S5)	0	3%	0
Sample 6 (S6)	0	5%	0
Sample 7 (S7)	0	0	1%
Sample 8 (S8)	0	0	3%
Sample 9 (S9)	0	0	5%

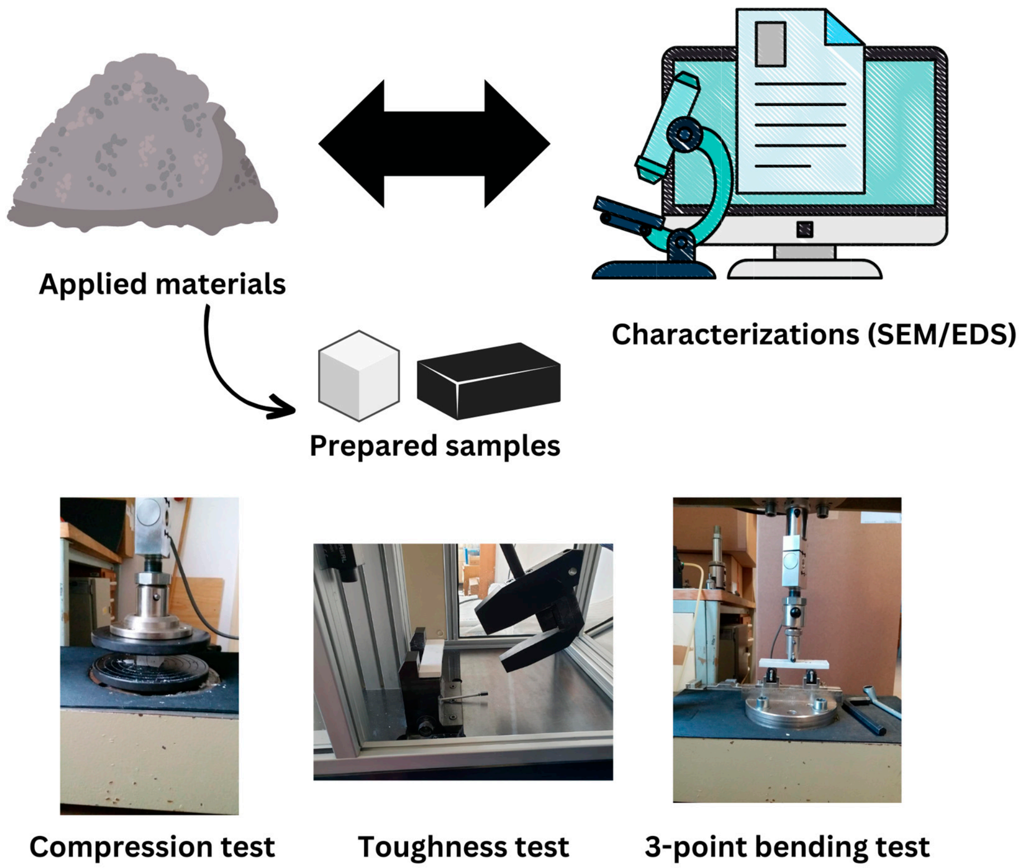


Figure 5. The experimental assessment of cement's performance in the investigation.

2.3. Instruments

The instruments used in this research are listed in Table 4. Some of the applied instruments are related to cement paste performance assessment and some others are connected to characterization of the prepared mixtures.

Table 4. The applied instruments in the study.

Device	Specification
Measuring Scale	Table Digital Accurate, Mettler-Toledo, s.r.o., Czech Republic
Mixer	KENWOOD XL TITANIUM, Great Britain
Vibrating Table	VSB-40 NS, Brio Harnice s.r.o., Czech Republic
Universal Testing Machine	Tira TEST 2300, Germany
Charpy Hammer LAB TEST	CHK 50J LABOR Tech, Czech Republic
SEM and EDS	VEGA3 TESCAN, Czech Republic

In the following, the results of the experimental practices that were carried out are modelled with linear regression models. Due to regression analysis, Excel software version 2016 was applied. In the process, the data are first categorized, and a fitting curve is produced from between different percentages of fillers and cement paste functions.

2.4. Environmental Impact (EIA) and Sustainability Assessment (SA)

In the present research, the created samples were first evaluated based on the toxicity of their existing elements based on immunological and epidemiological effects (Table 5). The EIA analysis in this study was implemented in MATLAB 2019b (Table 6).

Table 5. The toxicology of prepared sample scoring and their chemical formulas.

Additive	Formula	Elements	Toxic Level	Reference
FA	Si(6), Al(1), Na(2), H_2O(339)	Si, Al, Na, H, O	Al = 5, Si = 3, Na = 2, H_2O = 0, Li = 4, Mg = 4	[40,41]
BENT	(Na,Ca)0.33(Al,Mg)2$Si_4O_{10}(OH)_2 \cdot nH_2O$	Na, Si, Al, Mg, Ca, O, H		[42]
LAP	Na0.7Si_8Mg5.5Li0.3O20$(OH)_4$	Na, Si, Li, O, H		[43]

Table 6. The stages of EIA in this research.

No.	Stage	Descriptions
1	Calculation of Environmental Impact Scores	To assess the environmental impact of different mixtures, environmental impact scores are assigned to each chemical element present in the mixture. The following equation is used to calculate the environmental impact score (EIS) for a given element: EIS = ∑ (C_i × S_i) C_i represents the percentage of the element in the mixture. S_i represents the environmental impact score assigned to that element.
2	Sample Description and Calculation of Environmental Impact	For each sample, the environmental impact is calculated based on the chemical composition using the previously defined equation: EIS_sample_i = (C_Al × S_Al) + (C_Si × S_Si) + (C_Na × S_Na) C_Al, C_Si, and C_Na represent the percentages of aluminum, silicon, and sodium in the sample, respectively. S_Al, S_Si, and S_Na represent the environmental impact scores assigned to aluminum, silicon, and sodium, respectively.
3	Calculation of Total Environmental Impact	To obtain an overall measure of environmental impact for each sample, the individual environmental impact scores of aluminum, silicon, and sodium are summed up: Total_EI_sample_i = EIS_sample_i The Total_EI_sample_i represents the total environmental impact score for sample i.
4	Visualisation and Analysis	The obtained environmental impact scores are visualised using a bar graph. Each sample is represented on the x-axis, while the corresponding total environmental impact score is displayed on the y-axis. The bar graph provides a comparative analysis of the environmental impacts of different samples.

After quantifying the environmental impacts (EIs), the nine prepared samples (Table 3) are evaluated with respect to sustainability criteria, including economic and EI performance criteria. This assessment is based on ranking by using the OWA method in three stages described in Table 7. The ranking of criteria in the OWA method is based on the seven linguistic terms including very optimistic, optimistic, fairly optimistic, neutral, fairly pessimistic, pessimistic, and very pessimistic [44]. Note that the cost of LAP is the highest, followed by FA and then BENT, based on the prepared samples in Table 2 and market data.

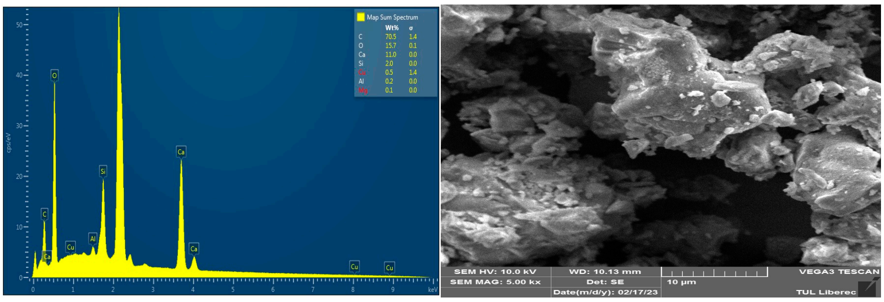
Table 7. The computation of OWA in this study.

Stage No.	Formula	Description	Reference
1	$F(a_1, a_2, \ldots, a_n) = \sum_{i=1}^{n} wibi$	In the given equation, the evaluations of alternative "a" with respect to "*n*" criteria are represented by the variable "a_i". It is mentioned that the inputs of the operator should be ranked in descending order. Therefore, "bi" represents the "i-th" largest element in the set of inputs "$a_1 \ldots n$". To calculate the associated weights of the OWA operator, the coefficients "wi" are used. These weights satisfy the conditions of being between 0 and 1, and their sum equals 1 (w = 1).	[44]
2	$wi = Q\left(\frac{i}{n}\right) - Q\left(\frac{i-1}{n}\right)$	Computing the weights	
3	$Q(r) = r^{\alpha}$	In the Q (r) equation, "r" is defined as the rank of "i" among "n" elements, with "i" ranging from 1 to "n". The coefficient "α" represents the optimism coefficient of decision makers (DMs), which has also been widely used to calculate linguistic quantifiers. Very optimistic: α = 0.0001 Optimistic: α = 0.1 Fairly optimistic: α = 0.5 Neutral: α = 1 Fairly pessimistic: α = 2 Pessimistic: α = 10 Very pessimistic: α = 10,000	

3. Results and Discussion

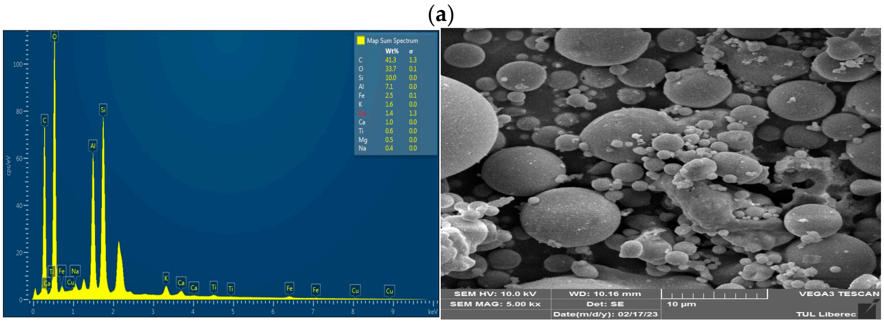
Figure 6 shows the results of EDS and SEM characterizations based on different raw materials and prepared samples.

Cement powder



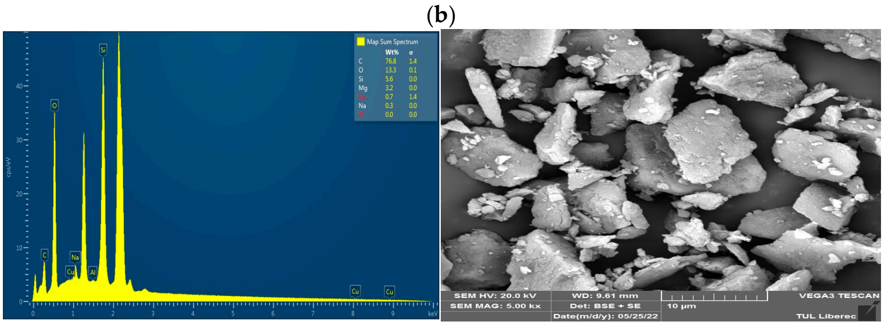
(**a**)

Fly ash



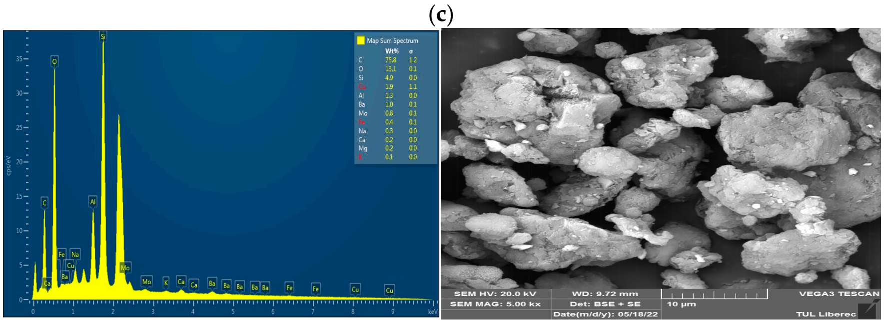
(**b**)

Laponite



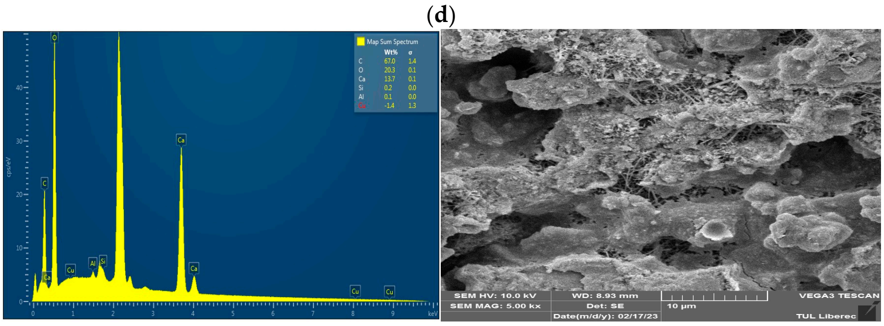
(**c**)

Bentonite



(**d**)

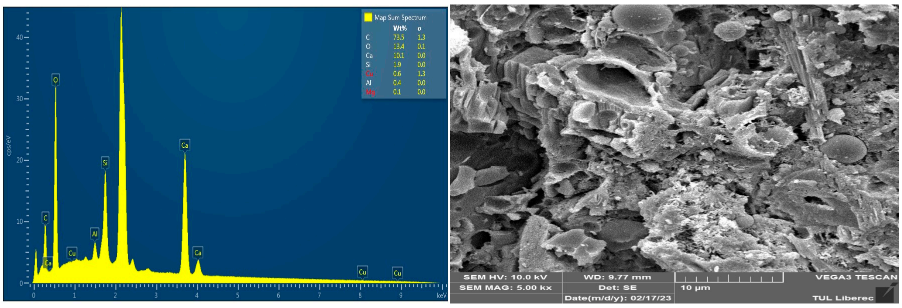
FA 5%



(**e**)

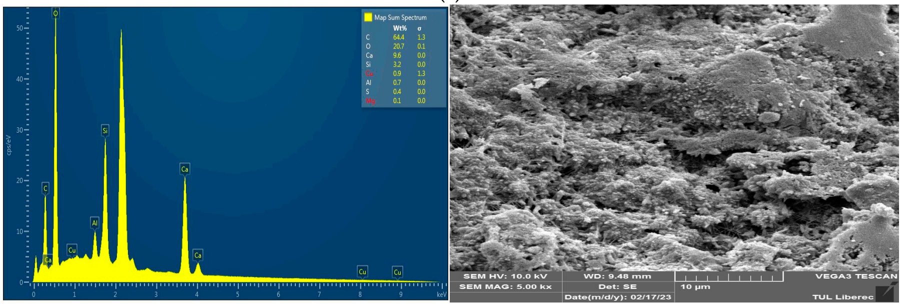
Figure 6. *Cont.*

FA 10%



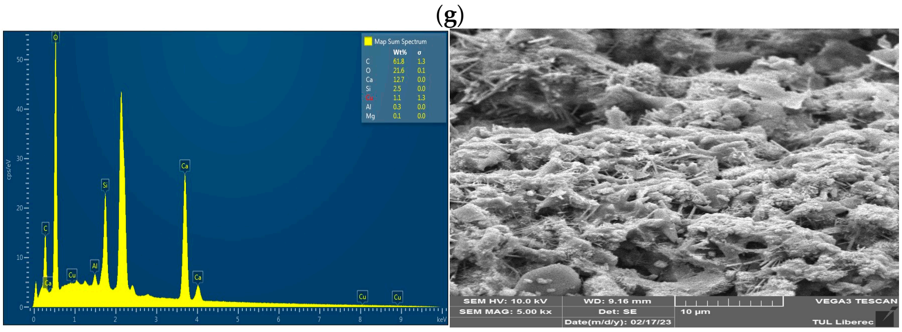
(f)

FA 20%



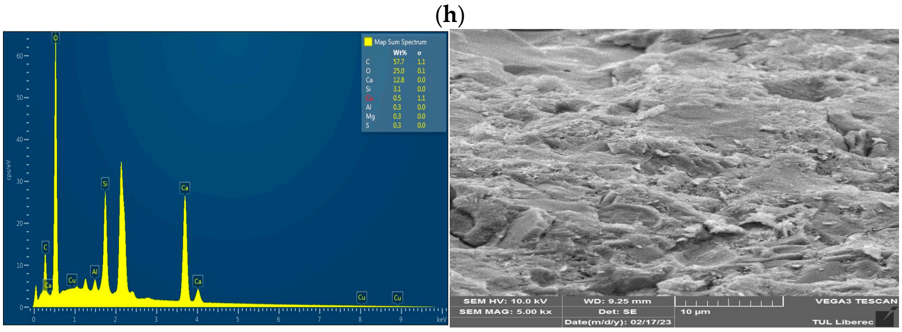
(g)

LAP 1%



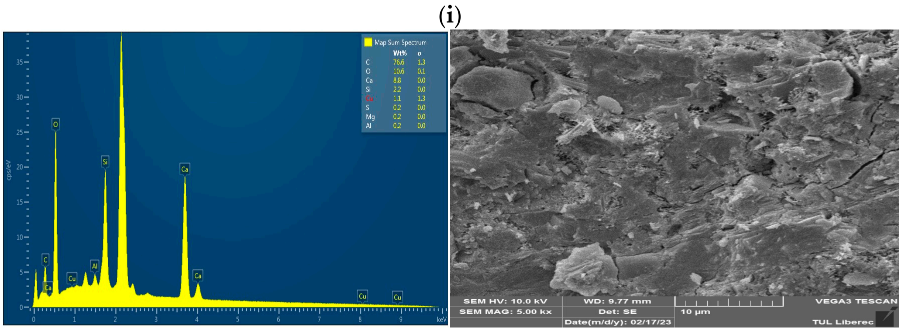
(h)

LAP 3%



(i)

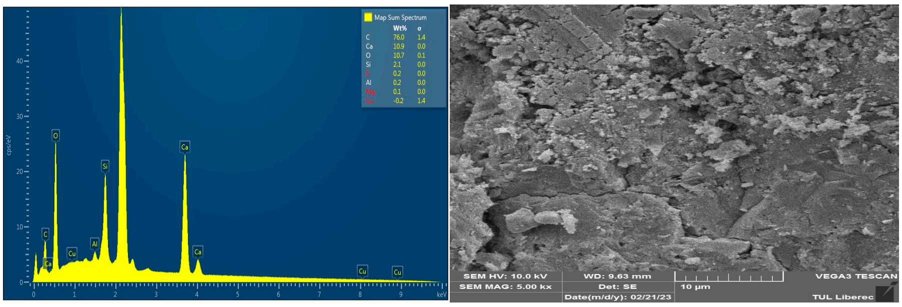
LAP 5%



(j)

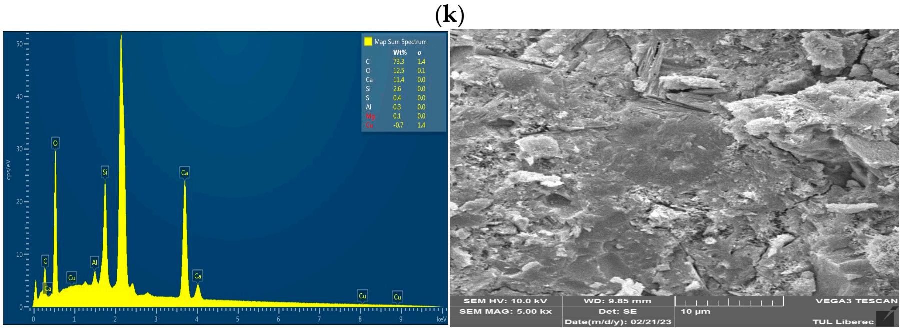
Figure 6. *Cont.*

BENT 1%



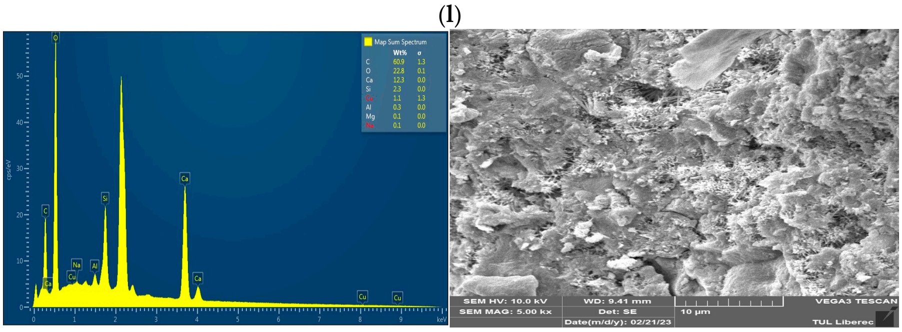
(**k**)

BENT 3%



(**l**)

BENT 5%



(**m**)

Figure 6. The results of characterization studies for the three cement paste additives with different percentages (**a**–**m**).

Figure 7 summarizes the results of the OFAT experiments. By comparing Figures 6 and 7, it can be seen that adding a higher percentage of fly ash in the mixture of cement paste would increase the percentage of C, O, Al, and Si elements and therefore increase the three-point bending value. However, increasing the aforementioned elements would considerably reduce the amount of toughness and compressive strength.

In comparing Figures 6 and 7, we can justify increasing the percentages of C, O, Al, and Si elements when adding more fly ash to the mixture of cement paste. These reasons can be attributed to several factors discussed herein. Firstly, the presence of fly ash in cement paste introduces additional reactive components into the mixture. Fly ash is a byproduct of coal combustion and contains a significant amount of silica (SiO_2) and alumina (Al_2O_3) [45]. These components react with the alkaline compounds in cement, such as calcium hydroxide, during the hydration process, forming additional calcium silicate hydrate (C-S-H) gel. The formation of C-S-H gel contributes to the overall strength and durability of the cementitious material [46]. Moreover, the increase in these elements can be linked to the pozzolanic reaction, which is a key mechanism associated with the incorporation of fly ash in cement paste. The pozzolanic reaction occurs between the fly ash particles and the calcium hydroxide present in the cementitious system. This reaction produces additional hydration products, including calcium silicate hydrate and calcium aluminate hydrate (C-A-H) gels. These gels fill the pore spaces within the cement paste, resulting in a denser microstructure and improved mechanical properties [47]. Additionally, the increase in the carbon and oxygen percentages can be attributed to the carbon content present in fly ash. Fly ash contains unburned carbon particles, which contribute to the overall carbon content of the cement paste when added to the mixture. The presence of carbon can influence the microstructure of the cement paste, affecting its mechanical properties [48]. The increased carbon content can enhance the binding of the cementitious materials, leading to improved three-point bending values. However, while the addition of fly ash and the subsequent increase in the mentioned elements can enhance certain properties, it is worth noting that there are trade-offs in terms of toughness and compressive strength. The incorporation of fly ash resulted in a decrease in toughness and compressive strength. This can be attributed to the dilution effect caused by the addition of fly ash, which leads to a reduction in the overall cement content and a decrease in the inter-particle bonding within the cementitious matrix [49,50]. Consequently, the material becomes more brittle and less resistant to applied forces.

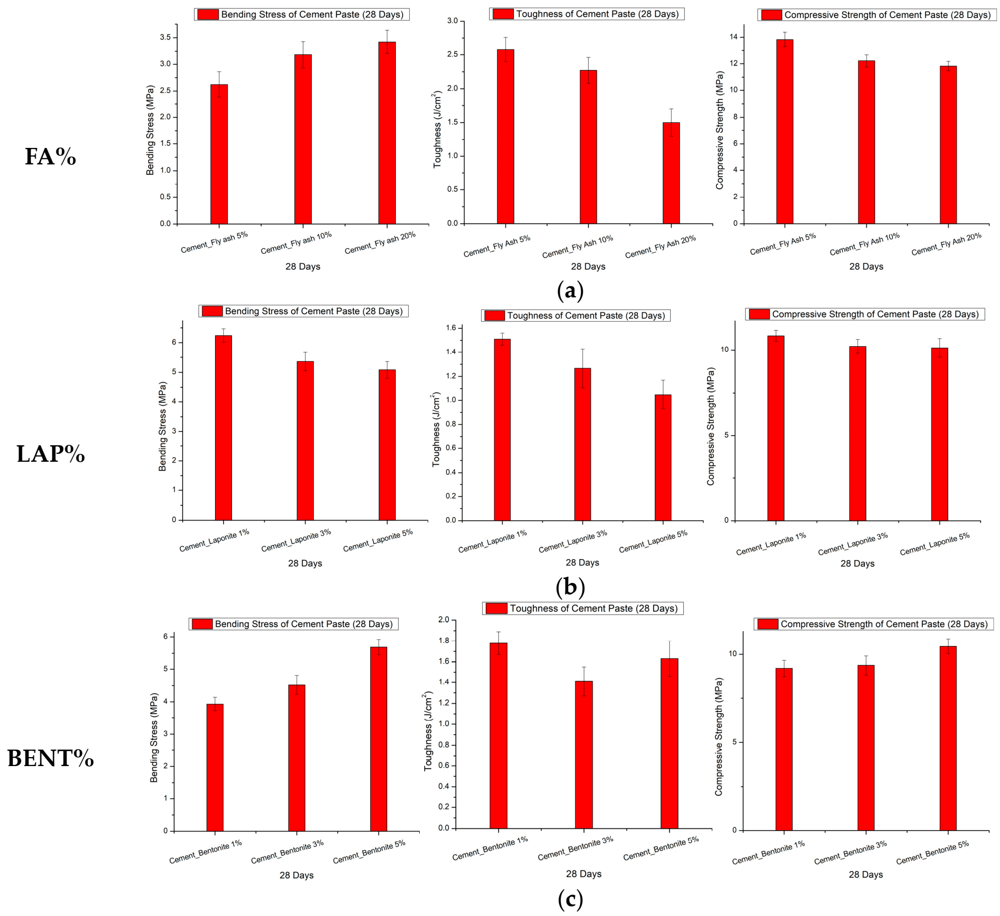


Figure 7. The results of the mechanical properties (bending stress, toughness, compressive strength) for the three cement paste additives: (**a**) FA%, (**b**) LAP%, and (**c**) BENT%.

By increasing LAP in the structure of the cement paste mixture, all functions (three-point bending, compressive strength, and toughness) can be reduced. Therefore, it can be concluded that as the C, Al, O, Si, Mg, and Na elements increase, the capability of cement paste is diminished. Therefore, upon elemental analysis of the addition of FA and LAP, it can be found that the Si, O, and C elements cause a decreasing in cement paste's specifications (three-point bending, compressive strength, and toughness). However, increasing Al has a positive effect on the three-point bending function. However, upon evaluation of BENT (main elements: C, O, Al, Si, Cu) three-point bending and compressive strength are increased; conversely, toughness is decreased. Thus, Cu and Al have positive influences on three-point bending, while only Cu has a direct relationship with compressive strength.

The presence of LAP can impact the hydration process of cement, which is essential for the development of strength in the paste [51]. The water associated with LAP, along with the water required for cement hydration, influences the availability of water molecules for the chemical reactions. Excess water from LAP can dilute the cementitious system, affecting the formation of stable calcium silicate hydrate (C-S-H) gel, which is responsible for the strength and durability of the cement paste. This dilution effect can lead to reduced three-point bending strength, compressive strength, and toughness. Furthermore, LAP contains carbon, which can directly affect the cementitious system. Carbon from LAP can react with calcium hydroxide ($Ca(OH)_2$) generated during cement hydration, leading to the formation of carbonates. This reaction can result in the consumption of calcium ions, which are essential for the formation of C-S-H gel. Consequently, the availability of calcium ions for C-S-H gel formation decreases, leading to a decrease in the overall strength of the cement paste. In the case of BENT, its chemical composition primarily comprises elements such as Si, Al, O, C, and Cu. These elements can interact with the cementitious system and influence its properties differently. The presence of Cu in BENT can have a positive effect on the mechanical properties of the cement paste. Copper ions can act as a catalyst in the hydration reactions, promoting the formation of a denser and stronger cementitious structure [52]. This can lead to an increase in three-point bending strength and compressive strength. On the other hand, Al in BENT can also influence the cement paste, particularly the three-point bending strength. Aluminum ions can react with the silicate compounds in the cement, leading to the formation of additional hydration products. These products can contribute to improved bonding and enhance the material's resistance to bending stresses. Also, when comparing the SEM images, it can be concluded that with increasing FA, LAP, and BENT, the porosity of cement paste is reduced. At the same time, all functions show different behaviors. Therefore, it can be concluded that the samples' strength outputs are independent of their porosities.

According to Figure 6, fly ash is a fine powder that is primarily composed of spherical particles. It is a byproduct of coal combustion in thermal power plants. The chemical composition of fly ash can vary depending on the composition of the coal burned, but it generally consists of silicon dioxide (SiO_2), aluminum oxide (Al_2O_3), iron oxide (Fe_2O_3), calcium oxide (CaO), and small amounts of other elements. The morphology of fly ash particles is typically irregular, with varying sizes ranging from a few micrometers to several tens of micrometers. The particles are often hollow and porous, giving them a light and powdery texture. The surface of fly ash particles can be rough and angular. Laponite is a synthetic clay-like material that belongs to the class of layered silicates. Its structure consists of a two-dimensional sheet of silica and magnesium ions, with water molecules located between the sheets. Laponite particles are disc-shaped, with a diameter typically ranging from a few nanometres to a few micrometres. These particles can stack together to form aggregates or gel-like structures in water or other polar solvents. Laponite has a high aspect ratio, meaning its width is much smaller than its length, resulting in a plate-like morphology. Bentonite is a type of clay formed from the weathering of volcanic ash. The structure of bentonite consists of individual clay platelets stacked on top of each other. These platelets have a layered structure, with each layer being composed of two silica tetrahedral sheets sandwiching an alumina octahedral sheet. The layers are held together by weak van der Waals forces, allowing them to slide and swell in the presence of water. Bentonite particles can vary in size, ranging from a few nanometres to several micrometres. They have a flake-like or needle-like morphology, with a high surface area due to the presence of numerous platelets. The interlayer spaces between the clay platelets can accommodate water molecules and other ions, giving bentonite unique swelling and adsorption properties.

It is well known that fly ash is a pozzolanic material that reacts with calcium hydroxide during hydration of cement to form additional cementitious compounds [53]. However, excessive amount of fly ash in the cementitious composites cannot fully participate in the pozzolanic reaction, thus resulting in a reduction of hydration products in cement-based composites. This can result in the reduction of the compressive strength [54]. It is important to note that higher compressive strengths are possible with higher fly ash content in cementitious composites when fly ash is mechanically activated, as shown in the outcomes of different studies [55–57]. Such studies are in accordance with the present investigation. The results of using LAP as a partial replacement in this study show that the compressive strength of cement pastes decreases with increasing LAP content. The possible reason for this is that LAP clay, being made up of very fine disk-like particles belonging to the Smectite group of phyllosilicates [43,58], possesses pozzolanic reactivity and can increase the C-S-H during the cement hydration process. Moreover, it can fill the voids, thus improving the packing of the cementitious system and increasing the compressive strength of the cement paste [59]. However, increasing LAP content causes agglomeration and also affects the water demand and workability of the cement, which can negatively affect the hydration procedure. Previous studies [60,61] reported similar results for compressive strength with different clays. The compressive strength of the cement paste with bentonite as a partial replacement for cement in this research work showed that a higher compressive strength value was achieved with 5% replacement of cement with BENT. This behavior of BENT in cement pastes may be due to the improved particle packing within the cement paste matrix. Furthermore, it may be due to the better binding and cohesion properties with increased hydration and cementitious products.

The outcomes of this study reveal that with increasing FA and BENT content, the three-point bending property of the cement paste also improved. This can be attributed to the better interfacial bonding between the FA/BENT particles and cement paste resulting in improved flexural strength [62]. However, in the case of LAP, three-point bending decreased with increasing content. This reduction in strength with increasing content of LAP may be because of the dilution effect of the LAP particles, which leads to poor participation in interfacial bonding.

Cement paste with FA/LAP/BENT in this study showed a decreasing trend with increasing contents of filler. In the case of FA, this behavior may be because of the increasing brittleness and reduced ductility of the cement paste, while the decreasing trend in the cement pastes with LAP may be attributed to the ineffective role of higher percentages of LAP in cement paste leading to poor toughness properties. Cementitious composites with partial replacement of cement by BENT firstly showed a decrease with 3% but at 5% showed some improvement. Cement paste with 3% showed a reduction of 20.8% in toughness compared to cement paste with 1% BENT, while cement paste with 5% BENT showed an improvement of 13.5% compared to cement paste with 3% BENT. This behavior may be due to the formation of cracks and their propagation due to the internal stresses within the cement pastes.

Using waste materials like fly ash and clays like laponite and bentonite as a partial replacement for cement is a promising area of study. However, there are certain issues and challenges to using these materials for the production of large volumes of concrete, including material variability, material availability for certain regions, technical issues for mixing and standardization at the site, and initial investments involved. In general, cementitious composites with these components can be used for non-critical components and for components where large-scale concreting is not required.

In the present study, a sensitivity analysis was conducted to precisely determine the effects of each additive on the mechanical properties of cement paste. This analysis was performed using a one-factor assessment approach. By varying one input variable at a time while keeping others constant, we assessed the impact of individual additives on specific targets, such as different strength parameters. This method allows us to identify the maximum sensitivity of the responses to input changes, providing a clearer understanding of how each additive influences the cement paste's mechanical performance. Future studies may benefit from a multifactorial approach to capture the interactions between variables for a more comprehensive analysis.

The results of regression statistical analysis for curve fitting of LAP, FA, and BENT against three-point bending strength, compressive strength, and toughness are summarized in Figure 8. The results show that after adding different percentages of FA in the mixture of cement paste, toughness and three-point bending strengths had acceptable ($R^2 > 82\%$) coefficients of determination. Likewise, the FA % addition had direct effects on three-point bending and had a reverse impact on both others. Figure 8 depicts that the R-Squared of all functions based on LAP is acceptable (more than 84%). However, the slopes of all functions (three-point bending, compressive strength, and toughness) have a negative one in the provided equations and show a reverse relationship with adding LAP to cement paste. The response of both three-point bending and compressive strengths in the case of BENT is acceptable ($R^2 > 84\%$) as they had a direct relationship with the additive. The reaction of BENT % and cement paste mixture vs. toughness was irregular, and the linear equation could not describe it.

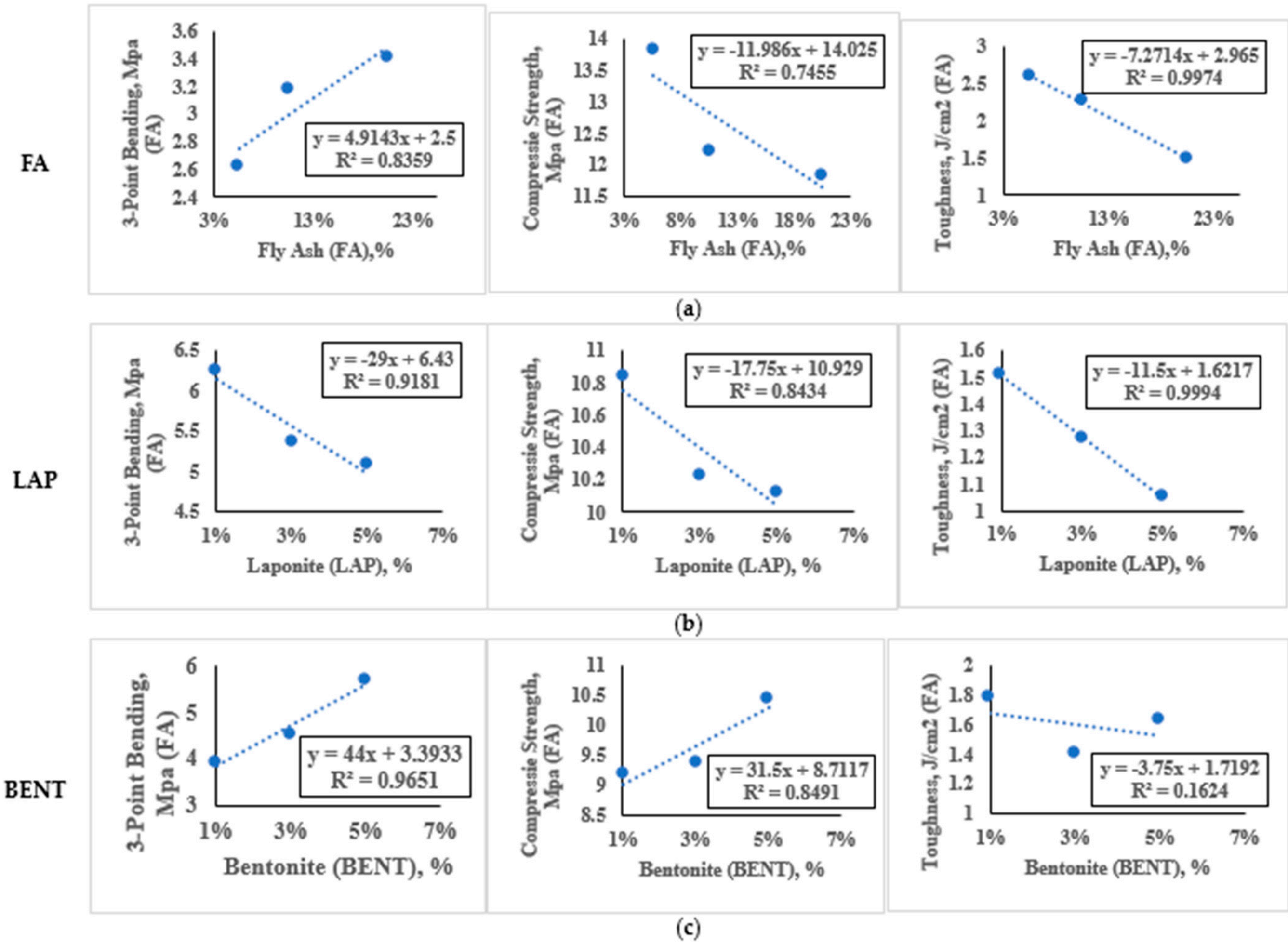


Figure 8. The results of regression statistical analysis for curve fitting in the present research (**a**–**c**).

Figure 8a demonstrates that FA% filler has the maximum effect on compressive strength (more so than the two others) because of its higher slope value in the obtained equations. In the next step, the higher relativity of FA% is linked to toughness (FA% vs. compressive strength: −11.98 > FA% vs. toughness strength: −7.2 > FA% vs. three-point bending strength: 4.9).

Regarding LAP% effects, it should be mentioned that in curve fitting computations, the slope values are LAP% vs. three-point bending strength: −29 > LAP% vs. compressive strength: −17.7 > LAP% vs. toughness strength: −11.5. Therefore, the maximum and minimum effects of LAP% addition to cement paste are connected to three-point bending strength and toughness, respectively. Compared with FA%, the order of the LAP% equation slopes was higher; thus, LAP% additives are more important than FA% in changing cement paste's specifications.

Finally, it is clear that the slope order of the BENT% equations in three-point bending and compressive strength is greater than both FA% and LAP% equations. Moreover, the BENT% effects on both mentioned functions were direct; however, the influences of both FA% and LAP% were reversed. Overall, similar to LAP%'s effects, in BENT%, the maximum slope (effect) and minimum one were related to three-point bending and toughness, respectively.

As a conclusion, FA significantly affects compressive strength, while LAP has the greatest negative impact on three-point bending and toughness. BENT improves three-point bending and compressive strength more effectively than FA and LAP, though it reduces toughness. Overall, BENT has the most positive influence on strength properties, while LAP has the most negative impact.

The outcomes of the EIA for nine samples are presented (Figure 9). The diagram illustrates that the presence of Al in the FA, and higher percentages of the additive used in the study resulted in the highest level of toxicity. Similarly, in the subsequent phase, varying percentages of BENT, due to the presence of Mg and Al elements, exhibited the most toxicity and EIs. Finally, based on its chemical structure, the least hazardous additive is associated with LAP.

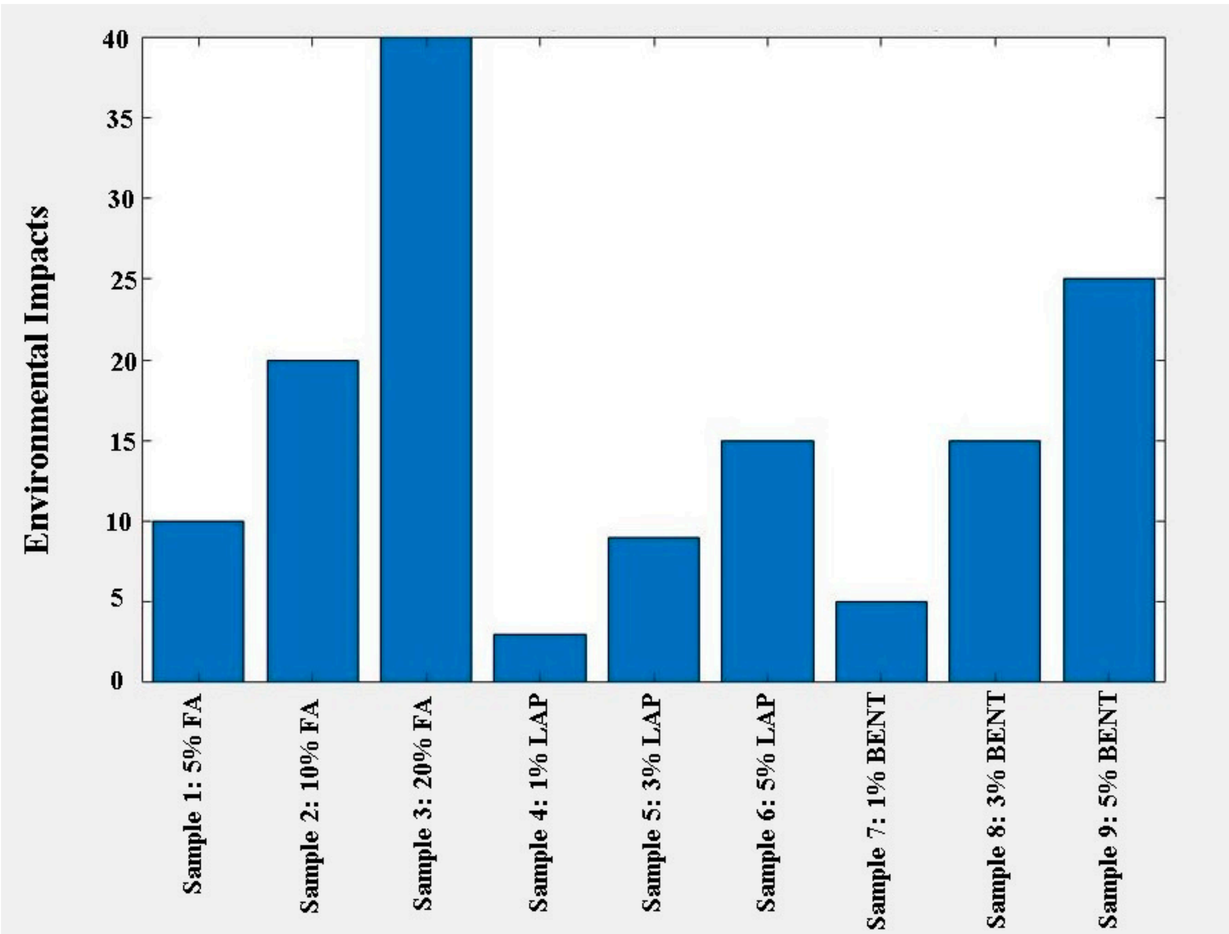
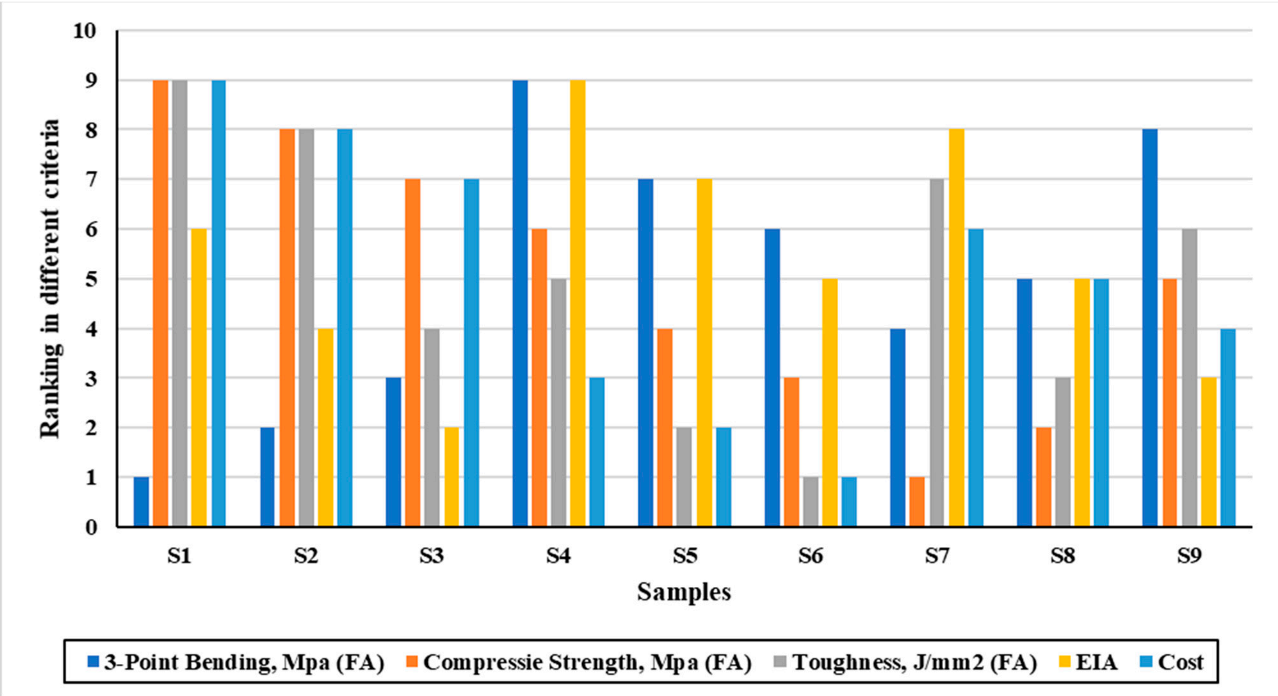
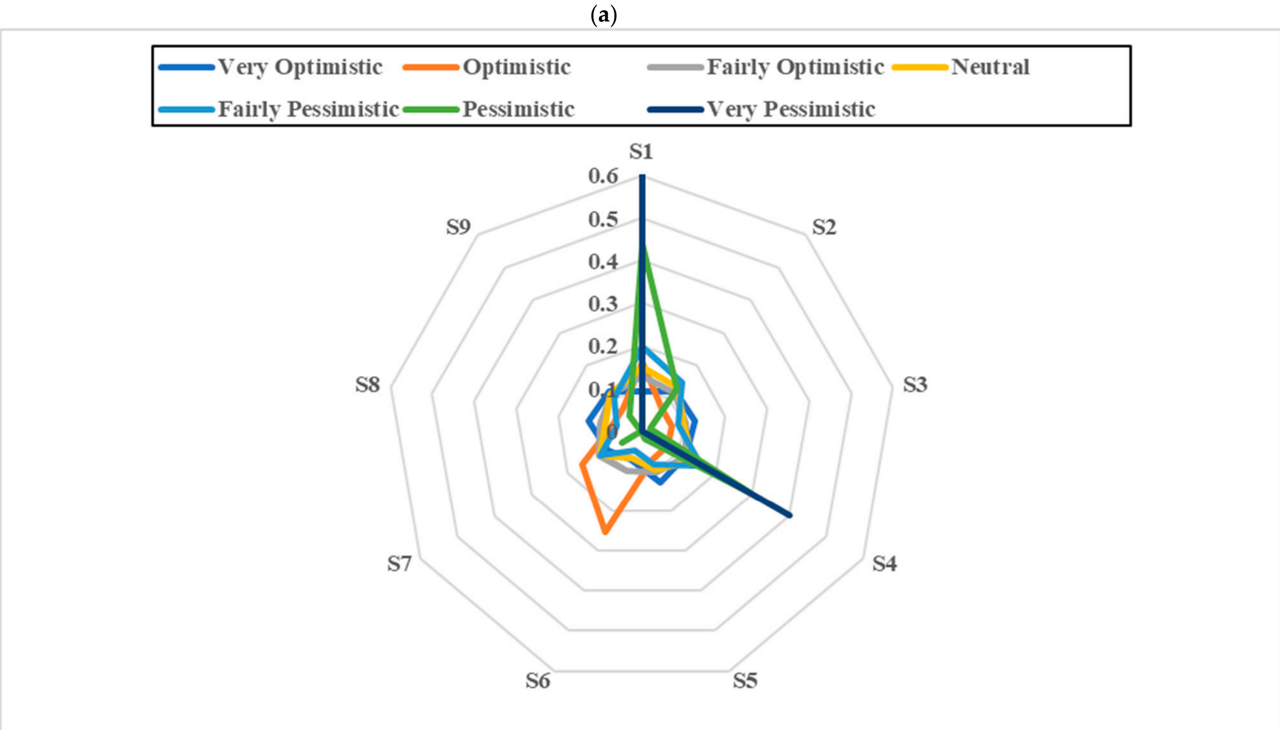


Figure 9. The outputs of EIA assessment of prepared samples in this research.

The results of the OWA ranking model are demonstrated in Figure 10. According to the scheme (Figure 10a,b), it can be concluded that based on SA indicators, from very pessimistic and neutral points of view (the views of managers), samples S1 and S4 should be selected for the construction preparation process. In both options, the lowest levels of FA (5%) and LAP (1%) are used as an additive to the structure of cement paste. Moreover, from a very optimistic selection perspective, S3 (with 20% FA, weight = 0.125), S5 (with 3% LAP, weight = 0.129), and S8 (with 3% BENT, weight = 0.126) are selected. Therefore, in an optimistic condition, all the materials can be applied based on the viewpoint of managers and decision makers. EIA is a useful tool, and in our case, 3% LAP is recommended.



(**a**)



(**b**)

Figure 10. The outcomes of OWA computations according to (**a**) sample rankings as per different performance/cost/EIA and (**b**) final OWA weights.

From a managerial perspective, it can be mentioned that adding higher percentages of FA increases elements such as C, O, Al, and Si, enhancing three-point bending strength but reducing toughness and compressive strength. Increasing LAP content reduces three-point bending, compressive, and toughness strengths due to dilution and excessive water demand. BENT improves three-point bending and compressive strengths but decreases toughness at higher concentrations. We should optimize FA and BENT content and limit LAP usage for balanced mechanical properties.

This study faced limitations, including the absence of FTIR tests, a limited number of samples, and financial constraints. Future research should incorporate FTIR analysis for better chemical insights, expand the sample range to include more industrial wastes for comprehensive analysis, and secure sufficient funding to enhance experimental scope and access to techniques.

4. Conclusions

This study analyzed and evaluated the performance of three green inorganic additives based on industrial wastes and clays (FA, LAP, BENT) for cement paste as a primary material in the construction industry. This study used some experimental practices to evaluate the effects of three percentage rates of FA, BENT, and LAP for the toughness, three-point bending, and compressive strength of cement paste. Furthermore, the process of sample preparation includes the assessment of the characterization of each sample via SEM and EDS tests. The statistical regression analysis proved the importance of each additive in different specifications of cement paste. EIA demonstrated that LAP is the safest studied additive to cement paste, and it can be selected based on SA analysis by OWA calculations given the very optimistic points of view of decision makers.

This study suggests evaluating the performance of LAP in lightweight concrete in comparison to FA and BENT from an economic point of view. The application of burned municipal solid waste ashes in the mixture of cement paste may also be attractive in some other research areas. Burnt municipal solid waste ashes can be utilized for the production of cementitious materials, and because of their high fineness, they can be partially replaced with cement, provided that they satisfy the requirements of the standards adopted [63]. The high lime content of burnt municipal solid waste ashes might enhance the cementitious properties of the prepared composites [64]. On the other hand, fresh and hardened properties of cementitious materials can suffer from the toxic compounds and elements that are present in burnt municipal solid waste ashes, for example, heavy metals, organic compounds, chlorides, and others [63]. To address this issue, the treatment of burnt municipal solid waste ash prior to its use in cementitious composites can give better results [65]. Finally, the circular economy assessment of the created mixtures on a larger scale can provide managerial insights for decision makers and is also a topic we suggest for future studies.

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Data Availability Statement: Data will be available on request.

Conflicts of Interest: The authors declare no conflicts of interest.

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