Selection of Novel Geopolymeric Mortars for Sustainable Construction Applications Using Fuzzy Topsis Approach

Manfredi Saeli 1, Rosa Micale 2, Maria Paula Seabra 3, João A. Labrincha 3 and Giada La Scalia 2,*

1 Department of Architecture, University of Palermo, 90128 Palermo, Italy; manfredi.saeli@unipa.it
2 Department of Engineering, University of Palermo, 90128 Palermo, Italy; rosa.micale@unipa.it
3 Department of Materials and Ceramics Engineering, Aveiro Institute of Materials, University of Aveiro, Campus Universitario de Santiago, 3810-193 Aveiro, Portugal; pseabra@ua.pt (M.P.S.); jal@ua.pt (J.A.L.)
* Correspondence: giada.lascalia@unipa.it

Received: 2 July 2020; Accepted: 22 July 2020; Published: 24 July 2020

Abstract: Construction is recognized as one of the most polluting and energy consuming industries worldwide, especially in developing countries. Therefore, Research and Development (R&D) of novel manufacturing technologies and green construction materials is becoming extremely compelling. This study aims at evaluating the reuse of various wastes, originated in the Kraft pulp-paper industry, as raw materials in the manufacture of novel geopolymeric (GP) mortars whose properties fundamentally depend on the target application (e.g., insulating panel, partition wall, structural element, furnishing, etc.). Five different wastes were reused as filler: Two typologies of Biomass Fly Ash, calcareous sludge, grits, and dregs. The produced samples were characterized and a multi criteria analysis, able to take into account not only the engineering properties, but also the environmental and economic aspects, has been implemented. The criteria weights were evaluated using the Delphi methodology. The fuzzy Topsis approach has been used to consider the intrinsic uncertainty related to unconventional materials, as the produced GP-mortars. The computational analysis showed that adding the considered industrial wastes as filler is strongly recommended to improve the performance of materials intended for structural applications in construction. The results revealed that the formulations containing 5 wt.% of calcareous sludge, grits, and dregs and the one containing 7.5 wt.% of calcareous sludge, grits, dregs, and Biomass Fly Ash-1 have emerged as the best alternatives. Furthermore, it resulted that the Biomass Fly Ash-2 negatively influences the structural performance and relative rank of the material. Finally, this case study clearly shows that the fuzzy Topsis multi-criteria analysis represents a valuable and easy tool to investigate construction materials (either traditional and unconventional) when an intrinsic uncertainty is related to the measurement of the quantitative and qualitative characteristics.

Keywords: construction; sustainable geopolymers; mortars; wastes valorization & reuse; fuzzy Topsis; multi criteria analysis

1. Introduction

The increasing pressure from worldwide environmental agencies to adopt an accurate waste management system is generating a growing interest in searching for sustainable alternatives to traditional disposal systems. The exploitation of non-renewable raw materials and the considerable production of wastes and greenhouse gases make the current global industrial system highly unsustainable.
In particular, construction is considered one of the most energy-intensive sectors and, for this reason, the criteria of sustainability and energy efficiency must be prosecuted to achieve the expected results foreseen by the recent European directives. Among the various construction materials, the cement industry is particularly critical. It is estimated that around one ton of CO\(_2\) is annually released in the atmosphere per ton of Ordinary Portland Cement (OPC) produced [1]. However, OPC exploitation is almost inevitable due to its easy and low-cost manufacture, large versatility, and easy application that make it a particular favorite for the construction of structures and infrastructures, especially in developing countries where enormous quantities of construction materials are required. As a result, developing alternative materials and cost-effective manufacture processes are becoming crucial to ensure a more sustainable industrial system worldwide.

In this work, the reuse of Kraft paper-pulp industrial wastes is analyzed in the manufacture process of novel sustainable geopolymeric mortars intended for applications in construction. The pulp-paper industry generates a great quantity of waste, resulting in around 11 million tons per year, only in Europe [2]. Some of these wastes show a high re-use potential and could be also be considered as by-products in other industrial sectors [3].

Geopolymer (GP), among the most popular eco-sustainable materials, has recently been playing a leading role worldwide. GPs are inorganic binders consisting of a solid alumina-silicate reactive source, frequently metakaolin (MK), which interacts with a solution—generally alkaline—to form a stable gel [4]. GPs are nowadays considered a sustainable alternative to OPC, as they show some interesting properties that make them suitable for various applications in construction. Among those, we recall structural elements, thermal-insulating and sound-absorbing panels, filters for wastewater decontamination, innovative multifunctional plastering, etc.

Several studies showed that MK can be substituted by other sources of alumina and silica [5–7], often deriving from many typologies of wastes such as blast furnace slag, fly ash, sludge, red mud, etc. Among these, a typology of Biomass Fly Ash (BFA-1), deriving from a Kraft paper-pulp industry, was used in this study to partially replace the MK (70 wt.% substitution), according to previous studies [8,9]. The Kraft pulping process is the most common industrial procedure to transform wood chops into wood pulp (fibers of nearly pure cellulose) that is the principal component of paper. This process entails several mechanical and chemical steps to treat wood with a hot mixture of water, sodium sulfide, and sodium hydroxide to break the bonds between lignin, cellulose, and hemicellulose. In the considered Kraft process, a large amount of solid and liquid wastes are generated [10,11].

In this work, GP-mortars were produced from the GP-binder by adding commercial siliceous sand (binder:aggregate—1:3). Five solid wastes, deriving from the same Kraft paper-pulp industry, were reused as filler with the primary aim of filling in the matrix pores and, consequently, increasing the mechanical strength of the GP-material. At the same time, the wastes reuse is maximized, improving the material sustainability, decreasing the exploitation of non-renewable raw materials and the overall manufacture cost. More particularly, the used wastes were: BFA-1 (already exploited as GP-binder precursor), another typology of Biomass Fly Ash, namely BFA-2, calcareous sludge (CalS), grits, and dregs.

In the literature, some recent works have studied new construction materials with multicriteria approaches, able to take into account, in the material formulation, not only the technical aspects, but also the economic and the environmental ones [12–14]. The need to consider these aspects adds controversial and often uncertain elements to the problem [15] and, for this reason, the selection of the decisional methodology is strongly influenced by the features of the problem itself. Numerous empirical [12,16] and theoretical [17,18] studies have shown that these computational techniques are optimal in the selection of new materials in which the above aspects must be considered simultaneously.

This paper aims to rank and select GP-mortars obtained reusing secondary raw materials from the paper-pulp industry that can be used in sustainable and innovative architectural design. This work demonstrates that the fuzzy Topsis approach can be considered a valuable multi-criteria support tool in the selecting process that, conversely, is not so immediate. The used technique starts from
defining some appropriate criteria of decision, evaluating their relative importance. The advantage of using the fuzzy Topsis mainly consists in allowing us to obtain a rank of the different alternatives expressed by means of fuzzy sets. In this way, the intrinsic uncertainty related to the measurement of the quantitative and qualitative criteria of unconventional materials can be considered. Moreover, the proposed method is compensative, meaning that the scores of the alternatives—with regard to the different criteria—are balanced. That does not happen, for instance, in the ELimination Et Choice Translating Reality (ELECTRE) methods [19]. Anyway, it must be pointed out that the obtained rank is strictly related to the specific considered application. This case study reports the analysis carried out on the formulations of mortars intended for structural application only.

At the best of our knowledge, the fuzzy Topsis approach has never been applied on any construction material and, moreover, on a geopolymeric material intended with specific uses in construction. That provides a fundamental advance in both the GP science and technology and the discipline of multicriteria analysis, providing a scheme of application, a selection of criteria and variables, and the related uncertainties, taken into account by the fuzzy approach.

2. Material and Methods

2.1. Materials

Based on former studies of the authors [8,9], a mixture of MK and BFA-1 (30/70 wt.%), activated by an alkaline solution made of sodium silicate (H₂O = 62.1 wt.%; SiO₂/Na₂O = 3.15; Quimialmel LDA, D40-PQ) and sodium hydroxide (ACS reagent, 97%/volume; Honeywell), was prepared to manufacture the GP-paste (binder). MK was purchased from Univar® as Argical™. Natural siliceous sand, furnished by Saint-Gobain Weber Portugal, was used as thin aggregate to produce the GP-mortar specimens. Based on Saeli et al. [8,20], the selected binder to aggregate ratio was 1:3.

The considered wastes—deriving from the Kraft paper-pulp industry—were: BFA-1 (another typology of biomass fly ash), calcareous sludge, grits, and dregs [21,22]. Biomass Fly Ash, typology 1, (BFA-1) derives from the main chemical recovery boiler in the biomass thermoelectric plant, and its average particle size is 60 µm; the Biomass Fly Ash, typology 2, (BFA-2) is generated in the auxiliary boiler of the steam cogeneration plant and presents an average particle size of 29 µm. Both the BFAs presented a negligible moisture content (~0.5 wt.%) and were used as received without performing any processes of drying, sieving, or milling. That improves the materials sustainability, as less energy is required.

Calcareous sludge (CalS) is an inorganic alkali residue made of calcite. It is produced from the chemical recovery circuit (White Bleach Clarifier) of the Kraft process as the molten inorganic salts are passed into the dissolution tank to be clarified. CalS was furnished in the form of powdery sludge characterized by an average particle size of 23 µm and an average moisture content of ~10.5 wt.% at mill site [23,24].

Grits (grits) also are an inorganic alkali residue mainly made of calcite. They are generated during the liquefied inorganic salts clarification by means of a lime slaker during the recovery of the chemical liquor that is used to digest wood. Grits were furnished in a granular form, with a particle size distribution ranging 1–12.5 mm and a negligible dust fraction (<2 wt.%), and an average moisture content of ~10 wt.% at mill site. This residue was investigated by the authors as coarse aggregate [25,26].

Dregs (dregs) are mainly made of sodium, calcium carbonates, and sulphides, and contains an organic fraction. They are generated by the separation of calcium carbonate and oxide during the clarification of the green liquor in the form of sludge, as furnished, with an average moisture content of ~25 wt.% and an average particle size of 11 µm [27,28].

All the considered wastes are classified as “non-hazardous” according to European Committee (2000) [29], then their reuse in construction is feasible. The most common method to handle these wastes is disposing of them in landfills. Nevertheless, according to European Committee (1975) [30] it should be
avoided, as it might generate a negative impact on the environment, resulting in groundwater pollution from toxic components leaching, soil contamination, and the emission of ugly odors [31]. Moreover, the economical disadvantages of this methodology should be considered due to the increasing costs of the disposal as consequence of the most recent regulations aimed at protecting the environment and the decreasing of available landfilling areas [32,33].

2.2. Wastes Treatments and Mortars Production

Prior to use, depending on the wastes’ nature, some treatments may be necessary as follows:

- **Washing:** Baths in distilled water may be necessary in the presence of salts, hazardous elements, or impurities (i.e., chlorides). Hot water would boost the action, but the cost will consequently increase. The number of required baths depends on the substances’ solubility and the possible materials loss (wt.%). In this work, the considered wastes were not washed.

- **Drying:** Is necessary in the case of high moisture content as it might influence the final GP-material molar ratios. In this study, drying was performed at 60 °C for 24 h (until reaching constant mass) in a conventional oven to remove the content of moisture. The manufacture efficiency could be improved if the waste is dried naturally at the mill site (i.e., under the direct sun exposure or in a ventilated space). That would lessen the employed energy for a more sustainable material manufacture.

- **Milling:** Is necessary to break the lumps or crush granular materials to ensure a better mixing uniformity and materials reactivity. In this study, it was performed in a ceramic mortar at lab scale. An industrial large-scale ball-milling would decrease the cost associated to this treatment.

- **Sieving:** Separation of the particles depending on the desired granulometry. In the case of the filler, the desired dimension is ≤63 µm. An industrial automated mesh strainer would reduce the timing of the operation and decrease the overall cost associated with manufacturing.

For each considered waste, the necessary treatment is specified in Table 1. The GP-mortars were prepared according to EN 998-2:2016 [34], the manufacturing steps are listed in Table 2.

Table 1. Treatments to be performed for each residue. BFA: Biomass Fly Ash; CalS: calcareous sludge.

| Treatment | Waste       | BFA-1 | BFA-2 | CalS | Grits | Dregs |
|-----------|-------------|-------|-------|------|-------|------|
| washing   | -           | 10 baths | - | -   | -   |
|           |             | 24 h/90 °C | yield 40% |      |      |      |
| drying    | 24 h/60 °C  | 24 h/60 °C | 24 h/60 °C | 24 h/60 °C | 48 h/60 °C |
| milling   | -           | -     | yes   | yes  | yes  |
| sieving   | -           | -     | yes   | -    | -    |

Table 2. Materials manufacturing at lab scale. MK: metakaolin.

| Phase | Operation | Length of Time | Intensity | Equipment |
|-------|-----------|----------------|-----------|-----------|
| 1     | dissolution of anhydrous sodium in distilled water | 24 h | 50 rpm | magnetic stirrer |
| 2     | mix MK with BFA-1 | 5 min | - | hand |
| 3     | mix sand with filler | 5 min | - | hand |
| 4     | homogenization of sodium silicate and hydroxide (1) | 5 min | 50 rpm | magnetic stirrer |
| 5     | mix of the alkaline solution (4) with the solid raw materials (2) | 9 min | 60 rpm | mixer |
| 6     | mix of the slurry (5) with the aggregate (3) | 1 min | 60 rpm | mixer |
| 7     | pour fresh slurry into molds | 2 min | - | hand |
| 8     | vibrate the molds | 2 min (+1 min set up) | set by industry | vibrating table |
Table 2. Cont.

| Phase | Operation                          | Length of Time | Intensity | Equipment |
|-------|------------------------------------|----------------|-----------|-----------|
| 9     | seal the molds with a plastic film | 2 min          | -         | hand      |
| 10    | set up to let specimens harden     | 24 h           | -         | -         |
| 11    | unseal and demold                  | 10 min         | -         | hand      |
|       | cure at ambient conditions (20°C, 65% RH) | 27 days     | -         | -         |

N.B. Prior to materials manufacture, the wastes may be treated.

2.3. Hypothesised Applications in Construction

GP-mortars can be used in a variety of applications in construction that may necessitate different requirements in terms of engineering performance, environmental impact, admissible load, cost, durability, etc.

The first step of the proposed method is the selection of the target possible applications and the specification of the characteristics required to optimize the GP-formulations. For this reason, four different applications have been initially selected for the novel GP-mortars:

- **Structure**: GP-mortars can be used to manufacture supporting (structural) elements such as pillars, beams, walls, preformed ashlars, bricks, etc.
- **Insulation**: GP-mortars can be used to manufacture technical elements (i.e., panels) useful to reduce the thermal exchange through the building envelope.
- **Internal partitions**: GP-mortars can be used to build technical elements useful to divide the inner space (internal walls).
- **Finishing**: GP-mortars can be used to realize surface coatings (i.e., plaster) or decorative elements.

These selected applications have been hypothesized in order to cover a vast range of possible uses in construction.

2.4. Evaluation Criteria

Generally, several criteria may be considered to optimize the formulation of a material depending on a specific application \[35,36\]. Subsequently, the GP mixes were related and analyzed on the basis of a selection of properties. Therefore, this methodology is used to optimize the GP blends analyzing some important categories such as materials’ properties (fresh and hardened states), economic, environmental, and safety performance, that are presented in Figure 1.

The selected criteria are defined as follows:

- **Workability** \([\text{cm}]\) returns the consistency of the fresh mortar and indicates the material attitude to be homogenously mixed and conveniently placed. It is related to the slurry properties and the specific considered application. In this study, workability was estimated by flow table test, according to EN 1015-3:1999 \[37\].
- **Bulk density** \([\text{kg/m}^3]\) was calculated geometrically on the specimens cured for 28 days. The given value is the average from three.
- **Uniaxial Compressive Strength** \([\text{MPa}]\) (UCS) indicates the break point at compression and was determined according to EN 998-2:2016 \[34\]. A universal testing machine (Shimadzu, AG-25TA), equipped with a 250 kN load cell running at 0.5 mm/min displacement rate, was used. The mean values are calculated from three tests, performed at 28 days of curing.
- **Axial Strain** \([\%]\) is the strain at rupture during the compressive strength test (cf. former bullet point) and was calculated as the quotient of the displacement at maximum strength and the initial length of the specimen.
- **Water Absorption** \([\%]\) indicates the quantity of water that the specimen can absorb once immersed in water. The weight variation (\(\Delta P/P\%\)) was determined using the Archimedes principle by
immersing the dried specimens in distilled water for 24 h after reaching constant mass. Three tests were performed to calculate the mean values on the specimens cured for 28 days.

- Thermal transmittance [W/m²·K] is the rate of the heat transfer through the specimen and was calculated using a spectrometer following the ISO 6946:2017 [38].
- Specific heat [J/kg K] is defined as the heat quantity supplied to a material given mass to induce a unit change in its temperature. It is calculated using a calorimeter.
- Cost [€/ton] was calculated considering the different percentages of waste used in the GP mixes. It represents the disposal cost related on each different waste.
- The Environmental Impact (EI) was measured using the Life Cycle Assessment (LCA) [qualitative] methodology proposed Kurda et al. in [13]. The EI was estimated basing on the non-renewable energy (PE-NRe) and the Global Warming Potential (GWP). This criterion was evaluated with the qualitative scale as used in [13].
- Toxicity [qualitative] was considered regarding the safety aspect for human health [39,40]. In this paper, toxicity is defined as the dangerousness of handling the raw materials during the GP production. This criterion was evaluated with a qualitative scale.

![Figure 1](image_url) Categories used in the optimization procedure.

2.5. Fuzzy Topsis Technique

In this study, a multicriteria methodology was used to determine the correlations between the analyzed criteria (cf. 2.4 Evaluation criteria) and the different GP-mortar formulations. In order to consider the data imprecision, the fuzzy logic was then introduced. Hence, fuzzy numbers were used to express some of the aforesaid criteria; the membership functions were embodied by Triangular Fuzzy Numbers (TFNs). A schematic visualization is presented in Figure 2.
A Triangular Fuzzy Number (TFN) is denoted as a triplet \((a_1, a_2, a_3)\). The values \(a_1\) and \(a_3\) represent the lower and the upper bounds of a real number, whose weight (membership) is 0, while all
the numbers between \(a_1\) and \(a_3\) have a weight in the interval \([0–1]\) (membership function).

Some preliminary information on the relative importance of each criterion are required by the fuzzy Topsis methodology. Consequently, each considered criterion \(w_j\) was assigned a weight that represents its importance. In this study, the criteria weights have been evaluated by a panel of experts using the Delphi methodology [41]. The panel was composed by four university professors whose scientific interests are in the field of construction technology, materials, and architectural design; three professional engineers; and two technicians working in material science laboratories. The criteria weights have been defined for each selected application in order to optimize the GP mixes based on it as reported in Table 3. For some specific applications, the relative criteria were considered negligible.

### Table 3. Weights of each criterion for the different applications.

| Application       | Workability | Bulk Density | Compressive Strength | Axial Strain | Water Absorption | Thermal Transmittance | Specific Heat | Cost | LCA | Toxicity |
|-------------------|-------------|--------------|----------------------|--------------|------------------|-----------------------|---------------|------|-----|----------|
| Structural        | 0.115       | 0.132        | 0.136                | 0.136        | 0.125            | 0.000                 | 0.000         | 0.122| 0.122| 0.112    |
| Insulating panel  | 0.088       | 0.095        | 0.085                | 0.085        | 0.105            | 0.118                 | 0.118         | 0.098| 0.108| 0.101    |
| Vertical partition| 0.106       | 0.095        | 0.091                | 0.087        | 0.076            | 0.095                 | 0.099         | 0.114| 0.122| 0.114    |
| Finishing         | 0.147       | 0.099        | 0.000                | 0.000        | 0.129            | 0.099                 | 0.082         | 0.147| 0.147| 0.151    |

The first step of the Topsis methodology consists in building a fuzzy decision matrix \(\tilde{R}\), where the \(n\) rows represent the GP formulations and the \(m\) columns the considered criteria (Equation (1)). The value located at the intersection of a row with a column embodies the performance of a decision alternative according to a criterion.

\[
\tilde{R} = [\tilde{r}_{ij}]_{mn}
\]  

(1)

where \(\tilde{r}_{ij}\) directly represents the fuzzy values of the uncertain criteria, because the correspondent fuzzy numbers range in the interval 0–1.

On the contrary, for the crisp criteria, additional operations of normalization and fuzzification are necessary. Functions of linear normalization are applied to each criterion. At the same time, each obtained value was triplicated in order to represent the three vertices of the TFN. In this way, the related fuzzy number is obtained.

The second step of the fuzzy Topsis is constructing the weighted normalized fuzzy decision matrix \(\tilde{V}\). Each element of the normalized decision matrix \(\tilde{V}\) is multiplied by the weights \(w_j\) of the corresponding criteria (Equation (2)).

\[
\tilde{V} = \begin{bmatrix}
w_1\tilde{r}_{11} & w_2\tilde{r}_{12} & w_3\tilde{r}_{13} & \cdots & w_n\tilde{r}_{1n} \\
\vdots & \ddots & \vdots & \ddots & \vdots \\
w_1\tilde{v}_{m1} & w_2\tilde{v}_{m2} & w_3\tilde{v}_{m3} & \cdots & w_n\tilde{v}_{mn}
\end{bmatrix}
\]  

(2)
Afterwards, as the Topsis methodology suggests, the positive ideal solution Azimuth (A*) and negative ideal solution Nadir (A⁻) were identified. These solutions, that in our case were represented by fuzzy numbers, can be defined in different ways.

In the fuzzy context presented in this work, an ideal positive value $\vec{v}_j = (1,1,1)$ and an ideal negative value $\vec{v}_j = (0,0,0)$ were considered.

The third step of the methodology consists in the calculation of the positive and negative relative distances (Equations (2) and (3)). In this study, a fuzzy rectilinear distance has been employed, thus considering the fuzzy distance function as the sum of the differences of fuzzy homologous components (Equations (3) and (4)):

$$\vec{d}_i^+ = \sum_{j=1}^{n} d(\vec{v}_{ij}, \vec{v}_j) \quad i = 1, 2, \ldots, m$$

$$\vec{d}_i^- = \sum_{j=1}^{n} d(\vec{v}_{ij}, \vec{v}_j) \quad i = 1, 2, \ldots, m$$

The final step combines the two distances in order to obtain the relative closeness coefficient. The following Equation (5) was used:

$$\tilde{C}_i = \frac{\vec{d}_i^+}{d_1^+ + d_2^+ + d_3^+} = \begin{bmatrix} \frac{\vec{d}_1^+}{d_1^+ + d_2^+ + d_3^+}; \frac{\vec{d}_2^+}{d_1^+ + d_2^+ + d_3^+}; \frac{\vec{d}_3^+}{d_1^+ + d_2^+ + d_3^+} \end{bmatrix}$$

2.6. Numerical Application: GP Mortars Mix Design

In this work, a numerical example is reported on the use of the considered wastes generated in the Kraft pulp industry. These wastes were used to prepare the GP-mortars intended for applications in construction. In this paper, structural applications only (i.e., pillars subjected to axial compression) were considered.

The GP-mortar formulations were designed on the base of previous studies [9,20]. The binder, made of a mixture of BFA (70 wt.%) and MK (30 wt.%) activated by the alkaline solution, was admixed with the sand (binder to aggregate ratio equal to 1:3). Three different mixtures of the analyzed residues, used as filler, were added to the mortar’s slurry with the following quantities: 2.5, 5.0, 7.5, 10.0 wt.%. The produced specimens are detailed in Table 4.

To build the decision matrix, all the criteria related to fresh and hardened properties, except for the compressive strength, are reported as crisp values as well as the costs. UCS is, in fact, affected by a measurement error that can be directly translated through the use of fuzzy membership functions. As reported in Caleca [42], these data are transformed in the related TFN in the following way: The mean value $\mu$ corresponds to the triangle vertex ($a_2$), the upper bound ($a_3$) is calculated adding the error to the mean value, and similarly, the lower bound ($a_1$) is calculated by subtracting it. The values of the toxicity criterion were obtained by interviewing the panel of experts, as suggested by the Delphi described in Section 3. The experts’ qualitative judgements have been converted into fuzzy numbers, as reported in Table 5. More particularly, the employed linguistic scale was translated into the corresponding fuzzy membership functions. There, the first and the third numbers represent the corners at the base of the triangle, while the second number is the vertex (cfr. “3. Results and discussion”, Figure 3). Five linguistic variables—ranging between very low (VL) and very high (VH)—have been employed. A smaller number of values would have resulted in a less precise definition of the concepts expressed by the experts, while a more detailed scale would have not allowed to achieve the full agreement among them. The used scale approach directly derives from the Saaty’s scale [43].
Table 4. GP-mortars mix design.

| Waste                        | n. (id) | Total Filler [wt.%] | CalS | Grits | Dregs | BFA-1 | BFA-2 |
|------------------------------|---------|---------------------|------|-------|-------|-------|-------|
|                              |         |                     | Relative [wt.%] | Relative [wt.%] | Relative [wt.%] | Relative [wt.%] | Relative [wt.%] |
| Reference                    | 1       | 0.0                 | -    | -     | -     | -     | -     |
| CalS + Grits + Dregs         | 2       | 2.5                 | 33.33| 33.33 | 33.33 | -     | -     |
|                              | 3       | 5.0                 |      |       |       |       |       |
|                              | 4       | 7.5                 |      |       |       |       |       |
|                              | 5       | 10.0                |      |       |       |       |       |
| CalS + Grits + Dregs + BFA-1 | 6       | 2.5                 | 25   | 25    | 25    | 25    | -     |
|                              | 7       | 5.0                 |      |       |       |       |       |
|                              | 8       | 7.5                 |      |       |       |       |       |
|                              | 9       | 10.0                |      |       |       |       |       |
| CalS + Grits + Dregs + BFA-2 | 10      | 2.5                 | 20   | 20    | 20    | 20    | 20    |
|                              | 11      | 5.0                 |      |       |       |       |       |
|                              | 12      | 7.5                 |      |       |       |       |       |
|                              | 13      | 10.0                |      |       |       |       |       |

Table 5. Qualitative judgements and the corresponding fuzzy membership.

| Variable          | Membership Function |
|-------------------|---------------------|
| Very High (VH)    | (0.0, 0.00, 0.20)   |
| High (H)          | (0.20, 0.30, 0.40)  |
| Medium (M)        | (0.40, 0.50, 0.60)  |
| Low (L)           | (0.60, 0.70, 0.80)  |
| Very Low (VL)     | (0.80, 1.00, 1.00)  |

The same table was implemented to translate in fuzzy numbers the values of the LCA criterion. In particular, the intervals reported in Kurda et al. [13] were used to define the corresponding qualitative variable.

The criteria of water absorption and axial strain are to be minimized; the others maximized. The score of workability criterion is represented by means of a trapezoidal membership function. The maximum score (1) is achieved in the range of values 16–22 cm, which represents the most suitable values of slurry workability considering the target application [9, 44], whereas in the intervals [10–16] and [22–30] the score was calculated with the following Equations (6) and (7):

\[
x - 10 \quad \frac{6}{10 \leq x < 16} \tag{6}
\]

\[
30 - x \quad \frac{8}{22 < x \leq 30} \tag{7}
\]

For external values of the above-mentioned intervals, the score is equal to 0. Table 6 shows the criteria values related to the different GP-mortars formulations. Accordingly, with the fuzzy Topsis procedure, criteria have been normalized and fuzzified, as reported in Table 7. For this kind of application, the criteria weights have been set using the results reported in Table 3.
Table 6. Decision matrix.

| Waste Mix Design (id) | Uniaxial Compressive Strength [MPa] | Bulk Density [kg/m³] | Water Absorption [wt.%] | Workability [cm] | Axial Strain [%] | Cost [ø/ton] | Toxicity | LCA |
|-----------------------|-------------------------------------|----------------------|------------------------|------------------|------------------|--------------|---------|-----|
| 1 (20.75; 21.66; 22.57) | 1832 | 13.00 | 21.00 | 1.83 | 0 | VL | L |
| 2 (23.66; 24.87; 26.08) | 1865 | 11.18 | 20.50 | 1.94 | 0.087 | L | M |
| 3 (25.76; 26.76; 27.76) | 1852 | 10.58 | 18.50 | 1.72 | 0.174 | L | H |
| 4 (25.98; 26.42; 26.86) | 1819 | 10.57 | 17.25 | 1.71 | 0.261 | L | H |
| 5 (25.51; 26.09; 26.67) | 1810 | 10.45 | 14.25 | 1.63 | 0.348 | L | H |
| 6 (23.13; 23.62; 24.11) | 1854 | 10.28 | 19.00 | 2.15 | 0.123 | M | M |
| 7 (25.26; 26.37; 27.48) | 1862 | 10.24 | 18.00 | 1.94 | 0.246 | M | H |
| 8 (27.42; 28.40; 29.38) | 1867 | 10.16 | 15.50 | 1.73 | 0.369 | M | VH |
| 9 (25.10; 26.52; 27.94) | 1878 | 11.20 | 12 | 1.81 | 0.492 | H | H |
| 10 (25.50; 26.50; 27.50) | 1869 | 10.59 | 19.00 | 2.07 | 0.144 | H | H |
| 11 (27.21; 28.42; 29.63) | 1882 | 10.19 | 18.00 | 2.03 | 0.289 | H | VH |
| 12 (27.68; 28.69; 29.70) | 1888 | 10.01 | 15.00 | 1.91 | 0.434 | H | VH |
| 13 (28.52; 29.62; 30.72) | 1852 | 10.21 | 12 | 1.89 | 0.578 | VH | VH |

* values expressed in a qualitative scale (cf. par. 2.4 Evaluation criteria).

Table 7. Fuzzified and normalized decision matrix.

| Mix Design (id) | Economic Safety | Environmental Safety | Break Point | Bulk Density | Water Absorption | Workability | Shortening | Cost | Toxicity | LCA |
|----------------|-----------------|----------------------|-------------|--------------|------------------|-------------|------------|------|----------|-----|
| 1              | 0.68            | 0.28                 | 0.00        | 1.00        | 0.38             | 0.00        | 0.80      | 0.80 | 0.80     |
| 2              | 0.77            | 0.71                 | 0.61        | 1.00        | 0.60             | 0.15        | 0.60      | 0.40 | 0.50     |
| 3              | 0.84            | 0.54                 | 0.81        | 1.00        | 0.17             | 0.30        | 0.60      | 0.20 | 0.30     |
| 4              | 0.85            | 0.12                 | 0.81        | 1.00        | 0.15             | 0.45        | 0.60      | 0.20 | 0.30     |
| 5              | 0.83            | 0.00                 | 0.85        | 0.71        | 0.00             | 0.60        | 0.40      | 0.20 | 0.30     |
| 6              | 0.75            | 0.56                 | 0.91        | 1.00        | 1.00             | 0.21        | 0.40      | 0.40 | 0.50     |
| 7              | 0.82            | 0.67                 | 0.92        | 1.00        | 0.60             | 0.43        | 0.40      | 0.20 | 0.30     |
| 8              | 0.89            | 0.73                 | 0.95        | 0.92        | 0.19             | 0.64        | 0.20      | 0.00 | 0.20     |
| 9              | 0.82            | 0.87                 | 0.60        | 0.33        | 0.35             | 0.85        | 0.20      | 0.20 | 0.40     |
| 10             | 0.83            | 0.76                 | 0.81        | 1.00        | 0.85             | 0.25        | 0.20      | 0.20 | 0.40     |
| 11             | 0.89            | 0.92                 | 0.94        | 1.00        | 0.77             | 0.50        | 0.00      | 0.00 | 0.20     |
| 12             | 0.90            | 1.00                 | 1.00        | 0.83        | 0.54             | 0.75        | 0.00      | 0.00 | 0.20     |
| 13             | 0.93            | 0.54                 | 0.93        | 0.33        | 0.50             | 1.00        | 0.00      | 0.00 | 0.20     |
3. Results and Discussion

Figure 3 shows the results obtained with the fuzzy Topsis procedure, Table 8 the relative rank.

| Waste                        | n. | Total Filler [wt.%] | Group Rank |
|------------------------------|----|---------------------|------------|
| Reference                    | 1  | 0.0                 | 4          |
| CalS + Grits + Dregs         | 2  | 2.5                 | 3          |
|                              | 3  | 5.0                 | 1          |
|                              | 4  | 7.5                 | 2          |
|                              | 5  | 10.0                | 3          |
| CalS + Grits + Dregs + BFA-1 | 6  | 2.5                 | 4          |
|                              | 7  | 5.0                 | 2          |
|                              | 8  | 7.5                 | 1          |
|                              | 9  | 10.0                | 3          |
| CalS + Grits + Dregs + BFA-1 + BFA-2 | 10 | 2.5                | 4          |
|                              | 11 | 5.0                 | 4          |
|                              | 12 | 7.5                 | 2          |
|                              | 13 | 10.0                | 4          |

Preference rank was obtained considering the coefficient of closeness associated to each GP-mortar formulation. The triangles spread represents the amount of uncertainty. Consequently, when two triangles overlap, a weak rank can be still defined and the related uncertainty is expressed by the ordinate of the intersection between the two triangles. A wide area of overlapping can be explained by a high uncertainty that is, conversely, related to the rank itself. That ultimately means that the uncertainty in the input data is too much to define an absolute preference.
Results clearly show that the mortar formulations could be classified into four different groups (cfr. Table 8): The best formulations intended for structural application, included indifferently in the first group, are numbers 3 (CalS + Grits + Dregs – 5 wt.%) and 8 (CalS + Grits + Dregs + BFA-1 – 7.5 wt.%). That is in line with the laboratory results: Good workability, high mechanical strength, low axial strain, low water absorption. However, the triangle that represents its fuzzy score almost overlaps the triangle corresponding to the sample containing 7.5 wt.% of all the wastes (second group). That is due to a high mechanical resistance that is counterbalanced (fuzzy approach) by a higher axial strain and slightly lower workability. Conversely, the last group includes the worst GP-mortars formulations. It is observed that adding the BFA-2 generally decreases the material rank. Indeed, a part from formulation n. 12 (CalS + Grits + Dregs + BFA-1 + BFA-2 – 7.5 wt.%) that is included in the second group (due to the very high mechanical resistance), all the other formulations resulted in the lower part of the rank. Even though a very high mechanical resistance was measured, highly convenient for the considered application, the very low workability along with the high water absorption strongly reduce the materials performance, and consequently discourage their use for the selected application (structural). Among the worst formulations (group 4), the unmodified material (n. 1) is included showing—among the others—the lowest UCS and the highest water absorption. Indeed, these are fundamental parameters for the materials selection intended for real applicability. In any case, it is quite expectable that adding the wastes as filler is aimed at improving the materials overall performance. This outcome clearly demonstrates the convenience in the wastes’ reuse to manufacture novel sustainable GP materials for applications in construction.

4. Conclusions

This paper demonstrates that the fuzzy Topsis can be considered a valuable decision-making support tool in selecting novel GP-mortars for sustainable construction applications, in the specific case of considering both quantitative and qualitative criteria. This technique allows the decision maker to define some appropriate criteria of decision and to evaluate their relative importance, that is judged by a panel of experts (Delphi technique). This methodology ensures the employment of a structured procedure for the decision process, in such a context where the presence of multiple conflicting objectives allows us to select the best compromise rather than the best alternative. The decision-making process provides a solution which is strictly related to the specific applications. In the case study, we analyzed novel green mortars intended for structural application. The rank obtained by means of the fuzzy Topsis procedure was reported as a fuzzy set. This fact principally allows us to establish a general rank among the different considered formulations defining, at the same time, the level of confidence which in fuzzy terms is measured by the possibility value.

For the considered study, it is clearly shown that adding the proposed industrial wastes as filler is strongly recommended to improve the materials’ performance toward an efficient use in construction. The GP-mortars n. 3 (containing 5 wt.% of CalS + Grits + Dregs) and n. 8 (containing 7.5 wt.% of CalS + Grits + Dregs + BFA-1) have emerged as the best alternatives. Furthermore, it resulted that adding BFA-2 as filler in the structural mortar negatively influences the material performance, and the relative rank.

To conclude, the approach proposed in this study allows us to take into account the uncertainty related to qualitative judgments, as well as those that may exist in the measurement of quantitative parameters.

Further works foresee the application of the fuzzy Topsis methodology to other sets of construction materials, adding other criteria of evaluation to generalize its usage. Furthermore, other applications will be considered to make the presented approach a valid decision-making support tool useful for a vast range of situations.

Author Contributions: M.S. designed the experimental plan, produced the specimens, and characterised the material; G.L.S. designed the experimental plan and applied the Topsis methodology; R.M. applied the Topsis methodology; J.A.L., M.P.S., and G.L.S. were in charge of the scientific coordination. M.S. and G.L.S. drafted
the manuscript, all the authors were involved in the manuscript implementation. All authors have read and agreed to the published version of the manuscript.

**Funding:** Saeli M. thanks the Italian Ministry of Education, University and Research (MIUR) in the framework of the PON “Research and Innovation 2014-2020”, “AIM: Attraction and International Mobility”, Section 2—Attraction with D.D. 407 of 27/02/2018 co-financed by the European Social Fund—CUP B7419000650001—id project AIM 1890405-3, area: “Technologies for the Environments of Life”, S.C. 08/C1, S.D. ICAR/10, for supporting this work.

**Acknowledgments:** The project Smart Rehabilitation 3.0, Innovating Professional Skills for Existing Building Sector (2019-1-ES01-KA203-065657) co-funded by the Erasmus+ Programme of the European Union.

**Conflicts of Interest:** The authors declare no conflict of interest.

**References**

1. Schneider, M.; Romer, M.; Tschudin, M.; Bolio, H. Sustainable cement production—Present and future. *Cem. Concr. Res.* 2011, 41, 642–650. [CrossRef]
2. Monte, M.C.; Fuente, E.; Blanco, A.; Negro, C. Waste management from pulp and paper production in the European Union. *Waste Manag.* 2009, 29, 293–308. [CrossRef] [PubMed]
3. Van Beers, D.; Bossilkov, A.; Lund, C. Development of large scale reuses of inorganic by-products in Australia: The case study of Kwinana, Western Australia. *Resour. Conserv. Recycl.* 2009, 53, 365–378. [CrossRef]
4. Davidovits, J. *Synthesis of New High Temperature Geo-Polymers for Reinforced Plastics/Composites*. SPE PACTEC’79; Society of Plastic Engineers: Brookfield Center, NY, USA, 1979; pp. 151–154.
5. Samantasinghar, S.; Singh, S.P. Effect of synthesis parameters on compressive strength of fly ash-slag blended geopolymer. *Constr. Build. Mater.* 2018, 170, 225–234. [CrossRef]
6. Hu, W.; Nie, Q.; Huang, B.; Shu, X.; He, Q. Mechanical and microstructural characterization of geopolymers derived from red mud and fly ashes. *J. Clean. Prod.* 2018, 186, 799–806. [CrossRef]
7. Rajamma, R.; Labrincha, J.A.; Ferreira, V. Alkali activation of biomass fly ash–metaakolain blends. *Fuel* 2012, 98, 265–271. [CrossRef]
8. Saeli, M.; Novais, R.M.; Seabra, M.P.; Labrincha, J.A. Mix design and mechanical performance of geopolymer binder for sustainable construction and building material. *IOP Conf. Series: Mater. Sci. Eng.* 2017, 264, 12002–12011. [CrossRef]
9. Saeli, M.; Tobaldi, D.M.; Seabra, M.P.; Labrincha, J.A. Mix design and mechanical performance of geopolymeric binders and mortars using biomass fly ash and alkaline effluent from paper-pulp industry. *J. Clean. Prod.* 2019, 208, 1188–1197. [CrossRef]
10. Demir, I.; Baspinar, M.S.; Orhan, M. Utilization of kraft pulp production residues in clay brick production. *Build. Environ.* 2005, 40, 1533–1537. [CrossRef]
11. Nurmesniemi, H.; Poykio, R.; Perämäki, P.; Kuokkanen, T. The use of a sequential leaching procedure for heavy metal fractionation in green liquor dregs from a causticizing process at a pulp mill. *Chemosphere* 2005, 61, 1475–1484. [CrossRef]
12. Moretti, L.; Di Mascio, P.; Bellagamba, S. Environmental, Human Health and Socio-Economic Effects of Cement Powders: The Multicriteria Analysis as Decisional Methodology. *Int. J. Environ. Res. Public Heal.* 2017, 14, 645. [CrossRef] [PubMed]
13. Kurda, R.; De Brito, J.; Silvestre, J.D. CONCRETop—A multi-criteria decision method for concrete optimization. *Environ. Impact Assess. Rev.* 2019, 74, 73–85. [CrossRef]
14. Sciolitino, R.; Micale, R.; Saeli, M.; La Scalia, G. Multi criteria evaluation of a sustainable alkali-activated concrete. In Proceedings of the XXIV Summer School Francesco Turco, “Augmented knowledge, A new era of Industrial Systems”, Brescia, Italy, 12–14 September 2019; pp. 314–320.
15. Funtowicz, S.O.; Ravetz, J.R. *Uncertainty and Quality in Science for Policy*; Springer Science and Business Media: London, UK, 1990.
16. Jansen, R. *Multiobjective Decision Support for Environmental Management*; Springer Science and Business Media: London, UK, 1992.
17. Castells, N.; Munda, G. International environmental issues: Towards a new integrated assessment approach. In *Valuation and the Environment—Theory, Method and Practice*; O’Connor, M., Spash, C., Eds.; Edward Elgar: Cheltenham, UK, 1999; pp. 309–327.
18. Munda, G. *Multiple Criteria Evaluation in a Fuzzy Environment—Theory and Applications in Ecological Economics*; Physika Verlag: Heidelberg, Germany, 1995.

19. Roy, B. ELECTRE III: Un algorithme de classements fonde sur une représentation floue des préférences en présence de critères multiples. *Cah. CERO* **1978**, 20, 3–24.

20. Saeli, M.; Seabra, M.P.; Labrincha, J.A. Alkali-activated mortars for sustainable construction materials: Effects of binder-to-aggregate ratio and curing conditions. In Proceedings of the 226th International Conference on Science, Technology, Engineering and Management (ICSTEM), Helsinki, Finland, 17–18 September 2019; pp. 16–23.

21. Dimmel, D.R.; Shepard, D.; Perry, L.F.; Joachimides, T.; McDonough, T.J.; Malcolm, E.W. Alkaline Pulping of Wood and Lignin Model Compounds in Aqueous Dmso. *J. Wood Chem. Technol.* **1985**, 5, 229–246. [CrossRef]

22. *Characterization of Minerals, Metals, and Materials, Minerals, Metals & Materials Series*; Springer: Cham, Switzerland, 2017.

23. Bajpai, P. *Pulp and Paper Industry*; Elsevier: Amsterdam, The Netherlands, 2016.

24. Saeli, M.; Senff, L.; Tobaldi, D.M.; Carvalheiras, J.; Seabra, M.P.; Labrincha, J.A. Unexplored alternative use of calcareous sludge from the paper-pulp industry in green geopolymer construction materials. *Constr. Build. Mater.* **2020**, 246, 118457. [CrossRef]

25. Tobaldi, D.M.; Senff, L.; Tobaldi, D.M.; Seabra, M.P.; Labrincha, J.A. Novel biomass fly ash-based geopolymeric mortars using lime slaker grits as aggregate for applications in construction: Influence of granulometry and binder/aggregate ratio. *Constr. Build. Mater.* **2019**, 227, 116643. [CrossRef]

26. Saeli, M.; Senff, L.; Tobaldi, D.M.; La Scala, G.; Seabra, M.P.; Labrincha, J.A. Innovative Recycling of Lime Slaker Grits from Paper-Pulp Industry Reused as Aggregate in Ambient Cured Biomass Fly Ash-Based Geopolymers for Sustainable Construction Material. *Sustainability* **2019**, 11, 3481. [CrossRef]

27. Modolo, R.; Benta, A.; Ferreira, V.M.; Machado, L.M. Pulp and paper plant wastes valorization in bituminous mixes. *Waste Manag.* **2010**, 30, 685–696. [CrossRef]

28. Novais, R.M.; Carvalheiras, J.; Senff, L.; Labrincha, J.A. Upcycling unexplored dregs and biomass fly ash from the paper and pulp industry in the production of eco-friendly geopolymer mortars: A preliminary assessment. *Constr. Build. Mater.* **2018**, 184, 464–472. [CrossRef]

29. EC (2000). Commission Decision 2000/532/CE; modified by Commission Decisions 2001/118/CE of 16 January, 2001/119/CE of 22 January and 2001/575/CE of 23 July. *Off. J. L.* **2000**, 226, 3–24.

30. European Council Directive 75/442/EEC of 15 July 1975 on waste. *Off. J. L.* **1975**, 194, 39–41.

31. Saikia, N.; Kato, S.; Kojima, T. Production of cement clinkers from municipal solid waste incineration (MSWI) fly ash. *Waste Manag.* **2007**, 27, 1178–1189. [CrossRef]

32. Jordan, M.; Sánchez, M.A.; Padilla, L.; Céspedes, R.; Osses, M.; González, B. Kraft Mill Residues Effects on Monterey Pine Growth and Soil Microbial Activity. *J. Environ. Qual.* **2002**, 31, 1004–1009. [CrossRef] [PubMed]

33. Nurmesniemi, H.; Pöykiö, R.; Keiski, R.L. A case study of waste management at the Northern Finnish pulp and paper mill complex of Stora Enso Veitsiluoto Mills. *Waste Manag.* **2007**, 27, 1939–1948. [CrossRef] [PubMed]

34. EN 998-2:2016. *Specification for Mortar for Masonry—Part 2: Masonry Mortar*; European Committee for Standardization: Brussels, Belgium, 2016.

35. Chudley, R. *Building Construction Handbook*; Routledge: London, UK, 2016; ISBN 978-1138907096.

36. Dassori, E.; Morbiducci, R. *Costruire L’Architettura. Tecniche e Tecnologie per il Progetto*; Tecniche Nuove: Milan, Italy, 2011; ISBN 9788848122986.

37. EN 1015-3:1999. *Methods of Test for Mortar for Masonry. Determination of Consistence of Fresh Mortar (by Flow Table)*; European Committee for Standardization: Brussels, Belgium, 1999.

38. ISO 6946:2017. *Building Components and Building Elements. Thermal Resistance and Thermal Transmittance. Calculation Methods*; BSI British Standards: London, UK, 2017.

39. Kurda, R.; Silvestre, J.D.; De Brito, J. Toxicity and environmental and economic performance of fly ash and recycled concrete aggregates use in concrete: A review. *Heliyon* **2018**, 4, 611. [CrossRef]

40. Rodrigues, P.; Silvestre, J.D.; Flores-Colen, I.; Viegas, C.; De Brito, J.; Kurda, R.; Demertzis, M. Methodology for the Assessment of the Ecotoxicological Potential of Construction Materials. *Materials* **2017**, 10, 649. [CrossRef]

41. Delbecq, A.L.; Van de Ven, A.H.; Gustafson, D.H. *Group Techniques for Program Planning: A Guide to Nominal Group and Delphi Processes*; Scott, Foresmann and Company: Glenview, IL, USA, 1975.
42. La Scalia, G. Solving type-2 assembly line balancing problem with fuzzy binary linear programming. *J. Intell. Fuzzy Syst.*, **2013**, *25*, 517–524. [CrossRef]

43. Aiello, G.; Enea, M.; Galante, G.; La Scalia, G. Clean Agent Selection Approached by Fuzzy TOPSIS Decision-Making Method. *Fire Technol.*, **2008**, *45*, 405–418. [CrossRef]

44. Caleca, L. *Architettura Tecnica*; Flaccovio Dario: Palermo, Italy, 2005; ISBN 978-8877583048.

© 2020 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).