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# Ceramics for Building Structures Made from Contaminated Soils: A Fuzzy Logic Intelligence Approach to Circular Mining

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**Abstract:** Soils contaminated by mining activities are a major environmental concern, and to avoid this type of environmental impact, carrying out high-cost processes is necessary. For this reason, a solution is proposed in this study in order to eliminate the soils contaminated by mining activities and, in turn, prevent the soil's contaminating elements from causing harm. All this is achieved by using contaminated soils as raw materials for the production of ceramics for bricks. For this purpose, the materials were initially characterized physically and chemically, and different ceramic test pieces were manufactured with different percentages of clay and contaminated soil, subsequently determining the physical properties and the leaching of toxic elements. In this way, it was possible to evaluate, via innovative data mining and fuzzy logic techniques, the influence of the contaminated soil's contribution on the properties of ceramics. Based on this, it was possible to affirm that the contaminated soil incorporation negatively affects the physical properties of ceramics as well as the leaching of polluting elements. The ceramic formed by contaminated soil and clay has a lower compressive strength, and it is associated with lower linear shrinkage and lower density, as well as higher porosity and cold-water absorption. However, the addition of different percentages of contaminated soil (up to 70%) to clay created a ceramic that complied with regulation restrictions. Therefore, it was possible to obtain a sustainable material that eliminates environmental problems at a lower cost and that fits within the new circular mining concept thanks to fuzzy logic techniques.

**Keywords:** circular mining; mining waste; contaminated soil; ceramic; construction materials; fuzzy logic; data mining; structures

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## 1. Introduction

One of the main industrial activities that contribute to the progress and prosperity of society is mining [1]. This activity extracts essential resources such as water, metals, aggregates, and fossil fuels, among others [2]. As a consequence, it is an indispensable activity that must remain active and comply with environmental regulatory frameworks for the sake of the socio-economic development of any country [3,4]. However, historically, this has not always happened; therefore, mining activities sometimes produce significant impacts on the environment, leaving an undesirable legacy of abandoned mining areas in the form of waste rock dams, open pit lakes, and other facilities that form a notable environmental liability [5,6]. Therefore, in the past, these mining activities have generated enormous quantities of waste, and some are potentially toxic, which have had a very significant environmental impact on flora and fauna and even on surface and groundwater [7,8].

Society's greater environmental awareness has made it possible to develop new environmental regulations that are much more restrictive for mining activity in terms of sustainability; however, the production of mining waste is an obligatory environmental impact of this sector that cannot be reduced [9]. For this reason, several research studies have been and continue to be carried out in the field of circular mining, particularly with respect to the reuse of mining waste in the manufacture of new materials [10,11]. However, few research studies have been carried out on the sustainable reuse of important elements such as soil, which is usually contaminated by mining activities [12].

Soil acts as a natural filter in nature, retaining and dispersing toxic or harmful elements to other media. Moreover, it is a living and dynamic system that is constantly changing [13]. The toxic elements that the soil receives can come from different sources; however, mining activities result in the most substantial soil contamination due to metallic elements that have difficulty degrading via natural processes [14].

Usually, the soil around mining production areas is contaminated by the leachate from different waste produced by these activities [15]. This is because all waste produced during different phases of mining activities is deposited outdoors, and rain processes make the dissolution of certain metals in the waste and their transport to nearby areas possible [16].

The principal pollutants that can contaminate the soil are arsenic, cadmium, chromium, copper, mercury, nickel, lead, tin, titanium, and zinc [17]. These toxic elements are produced during mining activities in considerable concentrations as they are, in most cases, concentrated together with the elements of mining interest [18].

However, for soil to be considered contaminated, two fundamental processes must take place [19]. The first process is the mobility of toxic elements: in other words, the capacity of these chemical elements to be transferred to other media. The second process is the ability of the toxic elements to enter into the biological processes of the micro-organism, called bioavailability. Both processes are controlled by environmental regulations in mining activities so that the contaminated soil does not affect the flora [20], fauna, or even the environment [21]. However, decades ago, the treatment of mining waste and soil contamination was not considered, as there were no environmental regulations. Therefore, nowadays, there are different soils contaminated by mining activities that have a significant environmental impact on the environment and people; thus, they need to be treated with appropriate processes to avoid contamination [22].

In order to remediate the possible environmental effects that may be caused by contaminated soil, two main processes are carried out: decontamination and stabilization. The process of soil decontamination consists mainly of its removal for subsequent transport to stabilized landfills that prevent the mobility of existing toxic elements [23]. On the other hand, the stabilization of contaminated soil consists of the use of different techniques to prevent toxic elements in the soil from being transferred to other media or from being bioavailable to organisms [24]. Among the different techniques used for the stabilization of contaminated soils is vitrification, which consists of increasing the temperature of the soil to 1600–2000 °C in order to form a vitrified ceramic matrix that prevents the leaching of toxic elements. However, the main disadvantage of the different processes described above is the high economic cost involved [25].

Consequently, in this research study, a completely new treatment for contaminated soils is carried out, and it takes advantage of the advantages of traditional decontamination and stabilization processes. This is because the contaminated soil is removed during the decontamination process so that it can be used as raw material for the formation of ceramics. At the same time, potentially toxic chemical elements in the contaminated soil are encapsulated in the ceramic matrix, thus preventing their leaching and the production of environmental problems. Therefore, the fundamental advantage of the contaminated soil treatment detailed in this research study is that the soil is used as a raw material, reducing the cost of manufacturing ceramics and, consequently, avoiding the costs of traditional soil decontamination processes.

The use of brick ceramics as a material to house the contaminated soil and prevent environmental impact was selected for several reasons [26]. On the one hand, the construction sector requires large quantities of raw materials, with consequent environmental impacts. Therefore, the use of a waste product such as contaminated soil would lower the costs of brick production and reduce the environmental impact caused by the extraction of clay for brick production. On the other hand, the brick-manufacturing ceramic industry is located close to the contaminated soil, so transport costs would be cost-effective. In addition, the ceramic matrix formed by the clay with the addition of the contaminated soil would prevent the leaching of potentially toxic elements from the soil [27]. It is therefore an environmentally friendly solution framed within the so-called circular mining concept [28].

Thus, in the present research study, ceramics are developed with soil contaminated by mining activities in the mining district of Linares. For this purpose, the materials are initially evaluated by determining their chemical composition and compatibility. Subsequently, several families of specimens with varying percentages of clay and contaminated soil are formed to determine their physical and mechanical properties. Since the purpose of this research is to evaluate the retention of contaminants in the ceramic matrix, the leachates from the different families of specimens are analyzed to corroborate this retention.

In addition, and as a novelty of this research study, the data resulting from different tests are analyzed using fuzzy logic and data mining with MATLAB software. In this way, unlike classical statistics, it is possible to analyze the entire data cloud obtained via different tests and establish the relationships between different variables to achieve greater resistance to compression. In other words, and taking as a fundamental premise that what is desirable in a ceramic brick material is compressive strength, the application of fuzzy logic allowed determining the existence of a series of cause–effect relationships between the different physical properties of conformed ceramics and various percentages of contaminated soil and compressive strength. This method is, therefore, a completely new technique that has never been previously carried out in scientific research studies in the field.

Finally, the main objective of this research study is the experimental development of new ceramic materials using soil contaminated by mining activities, and the materials are capable of lowering the production costs of these traditional materials and, in turn, transforming an environmental liability (contaminated soil) into an economic asset (construction material) by adding different proportions of these soils to the final ceramic, thereby retaining the potentially polluting elements of the soil. For this, the cause–effect relationships between different physicochemical variables related to ceramics and compressive strength are analyzed, with the latter being the prevailing property used to determine the feasibility of producing ceramic bricks from clay and contaminated soil.

## 2. Materials and Methods

### *2.1. Materials*

This section details the origin, location, and general characteristics of the two materials used for the development of this research study, namely contaminated soil and clay. Samples of both materials were taken from the research area according to the procedures detailed in the UNE-EN 932-1 standard. The samples were subsequently dried at a temperature of 105 ± 2 °C. In this way, the humidity variable could be eliminated so that it would not disturb the final results. In order to evaluate whether it is possible to use these materials directly without having to carry out more costly preliminary work, these materials were not crushed, and there was no reduction in their particle size.

The main research materials are described below.

#### 2.1.1. Contaminated Soil

The contaminated soil is located in the mining district of Linares, Andalusia, Spain (38°5′42.68″ N 3°38′9.67″ W). More specifically, this soil is situated in the vicinity of an old smelting industry called "La Cruz" (Figure 1). This smelter, founded in 1830 and now abandoned, was established for the manufacture of lead products from ores extracted from adjacent mines. However, in more recent times, different ores were processed to obtain various products and not only lead. As a result, until the closure of this industry, enormous quantities of waste containing various toxic elements were generated, and these toxic elements can still be found today in the smelter itself or within the vicinity of the smelter.

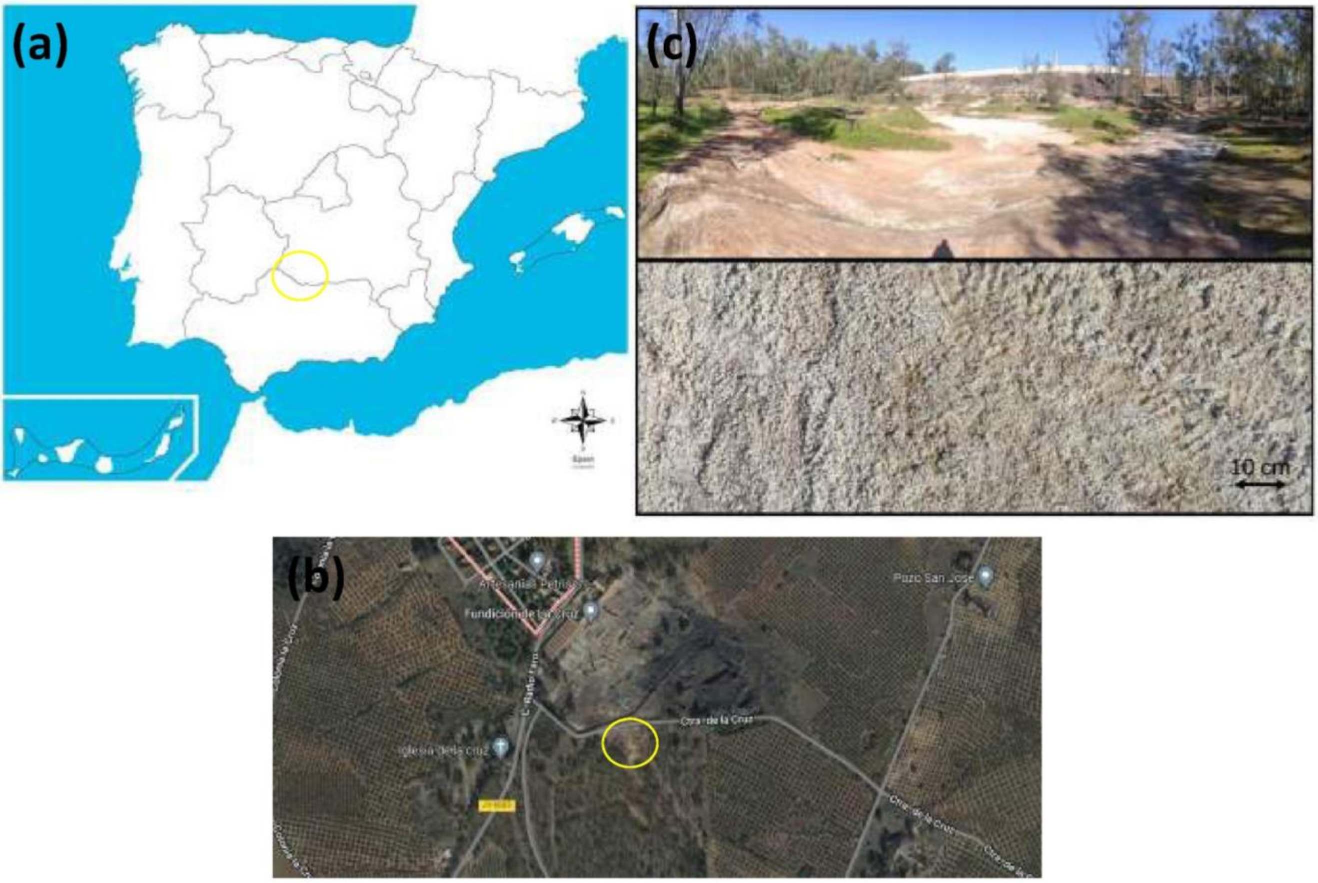


**Figure 1.** Location of the area in which the contaminated soil is located. (**a**) Map of Spain showing the location of the mining district of Linares. (**b**) Satellite image of the area in which the contaminated soil is located. (**c**) Image of the area of the contaminated soil and detail of the contaminated soil.

These types of waste, inherent to the process of concentrating lead and other metals for the manufacture of smelting products, appear to be responsible for the contamination of the soil in the study area adjacent to the installations. This is due to the fact that the waste located is outdoors and exposed to rainwater. Therefore, the action of rainwater on waste can cause the leaching of potentially toxic elements that are transported by water to lower levels. The adjacent soil, located at a lower level, seems to have received contaminated rainwater and has acted as a filter, as its plastic clay composition has retained the pollutants in the water. This contaminated soil (Figure 1) occupies an area of 300,000 $m^2$, representing a major environmental problem that no current study has focused upon; moreover, no solution has been proposed.

Soil samples were taken from the most superficial substratum, as the impermeable characteristics of natural soil make it possible to find most chemical contaminants on the surface. This sampling was carried out according to the UNE-EN 932-1 standard. These samples were used directly in the conformed ceramic after eliminating humidity, which means that no milling process was carried out.

#### 2.1.2. Clay

The clay used in this investigation corresponds to a clay known as red clay, which is usually used for ceramic materials in the construction sector. This clay is found in the vicinity of the mining district of Linares, so most brick factories are closely located in the area of contaminated soil. More specifically, the clay has been collected from Bailen, Andalusia, Spain (38°4′48.83′′ N 3°47′3.05′′ W).

The clay is usually extracted by mechanical means, transported to the industry, and then crushed to small particle sizes. Different ceramic materials are then formed and sintered at high temperatures. Therefore, the use of contaminated soil as a raw material not only avoids contamination caused by the soil but also eliminates the work of extracting the clay, which has a substantial environmental impact and a significant alteration of the area.

The collected clay was sieved to a particle size of less than 0.25 mm, thus obtaining a material similar to that used in the industry.

### *2.2. Methodology*

This section describes different fuzzy logic tests and techniques that are used to develop the methodology. This methodology will make determining the feasibility of using contaminated soil as a raw material for new ceramics possible. To this end, the materials were first physically and chemically characterized; then, several families of ceramic specimens were manufactured with contaminated soil and clay, and then the physical and mechanical properties of these specimens were determined, as well as the leachates of potentially toxic elements. Finally, thanks to the use of fuzzy logic and data mining techniques, it was possible to obtain the relationships between the different variables, obtaining a variety of combinations of contaminated soil and clay that made it possible to create marketable ceramic materials with particular characteristics.

#### 2.2.1. Physical and Chemical Characterization of Contaminated Soil and Clay

In order to physically characterize both contaminated soil and clay, the initial calculation of the particle density for both materials was carried out according to the UNE-EN 1097 standard. Subsequently, liquid and plastic limits were obtained in accordance with the UNE-EN ISO 17892-12 standard, with the aim of evaluating the suitability of these materials for ceramic manufacturing.

Next, the polluted soil and clay were submitted to a chemical characterization via the method of elemental analysis. This test method allows the determination of the percentage of elements present in the sample that are of lower atomic weight, such as carbon, nitrogen, hydrogen, or sulfur. The commercial equipment used for this purpose was the TruSpec Micro (TruSpec Micro, LECO, St. Joseph, MI, USA), manufactured by LECO. The elemental composition of both materials was determined using the commercial ADVANrXP+ (ADVANrXP+, Thermo Fisher, Waltham, MA, USA) via X-ray fluorescence analysis.

#### 2.2.2. Conformation of the Test Tubes and Tests

After verifying the compatibility between clay and contaminated soil for the mixing process, several sets of samples were created using both materials. To determine the physical and mechanical property variations caused by the addition of contaminated soil to the ceramic material, a set of conventional ceramics made only of clay was first manufactured. Then, different sets of ceramics were created with increasing percentages of contaminated soil at increments of 20% until the last set was made entirely of contaminated soil. It is important to note that each set consisted of 6 test tubes to ensure statistically valid results. The composition of each set of ceramics, including the percentage of clay and contaminated soil used, is presented in Table 1.

**Table 1.** Families of conformed ceramics with different combining percentages of clay and contaminated soil.

| Family | Clay % | Contaminated Soil % |
|---|---|---|
| S0 | 100 | 0 |
| S2 | 80 | 20 |
| S4 | 60 | 40 |
| S6 | 40 | 60 |
| S8 | 20 | 80 |
| S10 | 0 | 100 |

After establishing the families and determining the respective proportions of contaminated soil and clay, six test samples were produced for each family. This involved blending the two materials in their designated percentages and incorporating 10% water. The mixture was thoroughly stirred until a uniform composition of contaminated soil and clay, along with the appropriate moisture content, was achieved.

Next, the mixture was carefully transferred into a metal mold with internal dimensions: 60 mm length and 30 mm width. Once the mixture was poured into the mold, it was subjected to a pressure of 30 MPa, effectively increasing its density and achieving properties akin to those typically observed in the ceramic industry. Following this, the sample was dried for 24 h at a temperature of 105 ± 2 °C and was sintered individually for each of the six samples in each family. The sintering process involved maintaining a temperature of 950 ± 5 °C for a duration of one hour.

After the completion of the sintering process, physical testing was conducted on all specimens from different families. The initial test involved determining the linear shrinkage and weight loss of ceramics during sintering, following the UNE-EN 772-16 standard. This entailed measuring the dimensions and weight of the specimens before and after sintering to assess any variations that occurred.

Simultaneously, the cold-water absorption of ceramics from different families was assessed in accordance with the UNE-EN 772-21 standard. This test aimed to determine the ceramics' ability to absorb water when exposed to outdoor conditions.

Lastly, the bulk density and open porosity of test specimens were quantified to evaluate the achieved structural quality. This helped establish whether the addition of contaminated soil resulted in an open structure with numerous pores and lower resistance or a compact structure with fewer pores and higher resistance. The UNE-EN 772-4 standard was followed for this particular test.

Furthermore, as the ceramics developed in this study are intended for use in construction bricks, the compressive strength of the test pieces from each family was determined in accordance with the UNE-EN 772-1 standard. This allowed for an assessment of whether the utilization of contaminated soil in ceramic production resulted in materials with higher or lower resistance.

Moreover, since the primary objective of this research study is to reuse contaminated soil and prevent its environmental contamination, demonstrating that the ceramic matrix can effectively retain potentially toxic elements present in the soil and prevent their leaching is necessary. To achieve this, leaching tests were conducted on all ceramic families with varying percentages of contaminated soil using the TCLP method [29]. The ceramics were crushed to a size smaller than 10 mm, and the specified method was followed to obtain leachate from each ceramic family. Inductively coupled plasma mass spectrometry (7900, Agilent, Santa Clara, CA, USA) was employed to analyze the leachate. The resulting concentrations of toxic elements were then compared to the international limits established by the US-EPA [30]. This comparison enabled the determination of the maximum allowable percentage of contaminated soil that can be incorporated into ceramics without posing environmental concerns.

### 2.2.3. Data Mining and Fuzzy Logic

Fuzzy logic is a powerful tool for establishing cause–effect relationships between variables; unlike classical statistics, fuzzy logic allows obtaining semi-quantitative reasons that are much easier to interpret within a greater scope. In classical statistics, everything is governed by binary rules (yes–no; white–black); however, in fuzzy logic, an entire range of "decimals" between 0 and 1 and shadows of gray located between white and black can be discriminated. This technique has been widely used to define these relationships in mining scenarios with respect to the water–mining–environment trinomial [31]. In the field of geopolymers, this technique has also been implemented with strong results [32].

In this research study, the use of fuzzy logic has made it possible to establish the relationships that occur between the percentage of contaminated soil added to the ceramic and the physical and mechanical properties, in addition to the percentage of contaminating elements in the leachate. Furthermore, this technique allows the definition of an antecedent and a consequent, so it is possible to establish what must occur in the antecedents (initial variables) to obtain a determined value of the consequent (main variable). In this way, there is not a single solution to the problem but a range of solutions that allow, by simply modifying the percentage of contaminated soil and clay, obtaining ceramics with different physical, mechanical, and leachate properties.

In this particular study, MATLAB software was used for data analyses. The fuzzy logic analysis methodology is schematically represented in Figure 2.

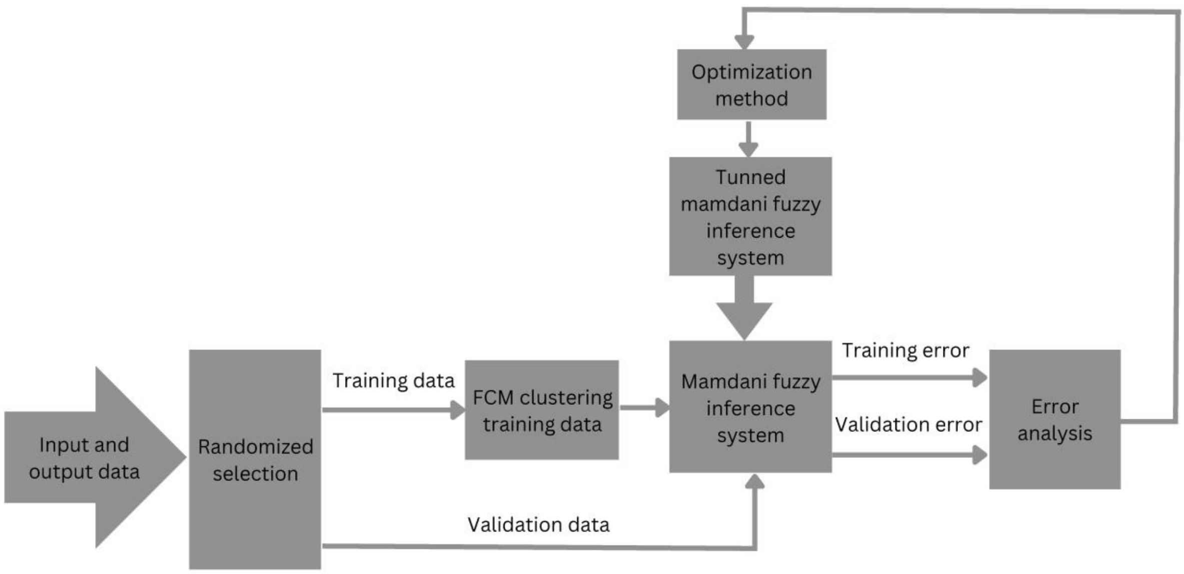


**Figure 2.** Flowchart of the fuzzy logic analysis methodology for MATLAB software.

All this can be explained more easily for the reader in the following example of Figure 3.

As observed in Figure 3, variables P1 and P2 (antecedents) directly influence variable S (consequent) and vice versa. Within the universe of discourse (which is made up of the range between the maximum and minimum value that each variable can acquire), for variable S (consequent) to acquire extreme–high values, variable P1 must acquire some intermediate–low results, and variable P2 must obtain intermediate–high results. Consequently, if variables P1 and P2 offered results other than those mentioned, consequent S would not acquire extreme–high values. All these values have a dimensionless character, which facilitates the system's modelling using large data masses in relation to heterogeneous variables in their dimension equations.

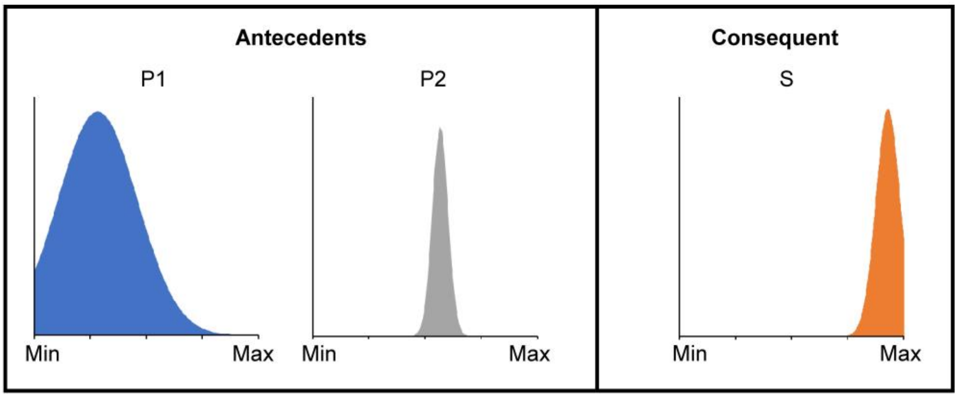


**Figure 3.** Graphical representation of fuzzy rules.

Therefore, as mentioned before, the potential of data analysis using fuzzy logic should be noted not only when different cause–effect relationships from classical statistics that relate variables to yes or no and 0 or 1 are established but also when dependency relationships can be established within a universe of discourse, which points to when relationships between the maximum and the minimum are obtained. In addition, the data analysis and representation with Gaussian curves offer more detailed information about the dependence of variables. This is because variable P1 has a Gaussian bell curve of greater amplitude than variable P2, so this reflects that there are many more values of variable P1 within its own universe of discourse that are capable of providing significant or maximized results of consequent S. However, the values of variable P2 are very concentrated within a range of the universe of discourse of the variable, so the data field is too small for obtaining high or maximum values of consequent S. This fact is mainly due to the shape and range of the represented Gaussian bell curve; this geometric shape depends directly on the central or maximum value, which corresponds to the mean, and its width on the data standard deviation: that is, the result's variation with respect to a central mean value. Consequently, a very wide Gaussian bell curve implies that the data variation or its standard deviation is high with respect to the universe of discourse. However, a very narrow Gaussian bell curve, as in the case of the P2 variable, implies that the data are very concentrated and that there is little difference between them [33,34].

Based on what was commented on, with fuzzy logic techniques, the various physical properties of contaminated soil and conformed ceramics were analyzed to evaluate the correlation of these variables with compressive strength, as this parameter is decisive for determining the viability of the commercialization of contaminated soil and conformed ceramic materials.

It should be noted that the results of the fuzzy logic analysis allow us to obtain a series of graphs in which the information is shown in a similar way to the previous example. In other words, the fuzzy logic graphs show antecedents and a consequent, establishing the values that each of the antecedents must take in order to obtain a determined value of the consequent. Thus, at a glance, the reader can quickly understand the cause–effect relationships between all variables [32].

Therefore, the proposed antecedents are as follows: linear shrinkage, weight loss, cold-water absorption, open porosity, bulk density, and chemical element concentrations (arsenic, lead, and barium) in the ceramic leachate and the contaminated soil percentage that is added to the clay to form the ceramic material, which in turn is the consequent compressive strength (MPa).

## 3. Results and Discussion

This section describes the test results mentioned in the methodology, as well as their partial discussion. In turn, as the main novelty of this research is framed within circular mining, all results were analyzed with fuzzy logic and data mining in an easily interpretable way, establishing the relationships between the different variables with a novel and complex methodology.

### 3.1. Physical and Chemical Characterisation of Contaminated Soil and Clay

The contaminated soil belonging to the Linares mining district had a density of 2.790 ± 0.066 g/cm$^3$. In turn, the plasticity index of the soil is 7.1 ± 0.1, which is lower than usual with respect to clay, as expected, but sufficient to make combinations of both materials in high percentages.

The chemical composition of the contaminated soil had a low percentage of lower atomic weight elements such as carbon, hydrogen, and nitrogen, resulting in a very low loss on ignition (2.94 ± 0.07). This reflects the fact that the analyzed soil has a low percentage of organic matter and hydrated compounds. The elemental analysis of the contaminated soil revealed a high percentage of silicon, aluminum, iron, and calcium and, to a lesser extent, potassium and sodium. Therefore, the analyzed soil comprises aluminosilicate. At the same time, percentages of 0.649 ± 0.032 of lead, 0.491 ± 0.50 of barium, and 0.142 ± 0.021 of arsenic were detected as potentially contaminating elements. These toxic elements in the contaminated soil must be evaluated after the manufacture of various ceramics to corroborate that their vitrification occurs inside the ceramic matrix.

The clay used in the present investigation had a particle density of 2.443 ± 0.063 g/cm$^3$, i.e., very similar to that of the contaminated soil. On the other hand, the plasticity index of the clay was 16.4 ± 0.6, which reflects the quality of this material for the shaping of ceramics intended for use as bricks. The percentage of lower atomic weight elements, such as carbon, nitrogen, and hydrogen, was low, resulting in a loss on ignition of 7.90 ± 0.25. This loss-on-ignition value, which is higher than that of the contaminated soil, reflects the composition of the clay, which is made up of hydrated compounds.

In turn, the elemental analysis of the clay reflected a high percentage of silicon and aluminum, obviously demonstrating that it is an aluminosilicate, with lower percentages of iron, calcium, potassium, and magnesium. Therefore, the chemical composition of the clay is ideal for its intended brick-making applications, showing no toxic elements such as lead, arsenic, and barium. Consequently, if these toxic chemical elements were present in the leachate of the different families of ceramics made of contaminated soil and clay, they would only come from the contaminated soil and never from the clay.

### 3.2. Conformation of the Test Tubes and Tests

The results of different physical and mechanical tests, to which the various families of specimens manufactured using different percentages of contaminated soil and clay were subjected, are shown in Table 2.

**Table 2.** Results of the physical and mechanical tests conducted using various families of ceramics made with clay and increasing percentages of contaminated soil.

| Contaminated Soil % | Linear Contraction % | Weight Loss % | Cold-Water Absorption % | Open Porosity% | Apparent Density g/cm$^3$ | Compressive Strength MPa |
|---|---|---|---|---|---|---|
| 0 | 4.90 ± 0.11 | 8.19 ± 0.19 | 4.3 ± 0.11 | 12.98 ± 0.34 | 2.23 ± 0.06 | 93.7 ± 2.47 |
| 20 | 2.38 ± 0.06 | 6.98 ± 0.16 | 7.08 ± 0.18 | 19.91 ± 0.53 | 2.14 ± 0.05 | 58.18 ± 1.54 |
| 40 | 0.56 ± 0.01 | 5.69 ± 0.15 | 8.54 ± 0.22 | 23.04 ± 0.59 | 2.14 ± 0.05 | 54.75 ± 1.45 |
| 60 | −0.53 ± 0.01 | 4.81 ± 0.12 | 9.3 ± 0.24 | 24.49 ± 0.65 | 2.07 ± 0.05 | 37 ± 0.95 |
| 80 | −1.01 ± 0.02 | 4.3 ± 0.11 | 9.97 ± 0.26 | 25.89 ± 0.71 | 2.05 ± 0.05 | 25 ± 0.66 |
| 100 | −1.73 ± 0.04 | 3.35 ± 0.08 | 12.32 ± 0.36 | 30.59 ± 0.72 | 1.89 ± 0.05 | 4.25 ± 0.12 |

As observed in Table 2, the average values of the evaluated physical properties of different families (made with clay and increasing percentages of contaminated soil up to 100%) are shown. It should be noted that these results will later be treated using fuzzy logic techniques; however, it should be mentioned that the standard deviation and standard error of the data for each family and variable in all cases are small. It is, therefore, an excellent data mass to analyze later using fuzzy logic, and the cause–effect relationships between the analyzed variables can be determined.

On the other hand, in order to corroborate that the potentially toxic elements of the contaminated soil are retained in the ceramic matrix, the TCLP test was developed, as mentioned in the methodology of [29]. Thus, it can be confirmed that the objective of this research, which is none other than to prevent contaminated soil from producing a significant environmental impact, has been achieved. For this purpose, the values of the concentrations of the polluting elements found in the ceramic leachate are compared with the limitations established by the international US-EPA organization [30].

These limits are indicated in Table 3.

**Table 3.** Maximum concentrations of metals or toxic elements in leachates according to the TCLP (US-EPA) method for construction materials.

| Metals | Maximum Admissible Concentrations in Leachates (ppb) |
|---|---|
| Pb | 5000 |
| As | 5000 |
| Ba | 100,000 |

The leachates obtained from the TCLP test of various families of ceramics, which are made up of different percentages of contaminated soil, reflected the concentrations shown in Table 4 with respect to the toxic elements restricted by US-EPA limitations.

**Table 4.** Results of the leaching tests of different families of ceramics formed by clay and soil contaminated by mining activities: for arsenic, lead, and barium elements.

| % Contaminated Soil | Arsenic (ppb) | Lead (ppb) | Barium (ppb) |
|---|---|---|---|
| 0 | 1.25 ± 0.03 | 0.98 ± 0.02 | 0.73 ± 0.02 |
| 20 | 399 ± 10 | 229 ± 11 | 121 ± 3 |
| 40 | 843 ± 22 | 638 ± 16 | 145 ± 4 |
| 60 | 1205 ± 32 | 1043 ± 31 | 179 ± 5 |
| 80 | 1703 ± 45 | 1348 ± 36 | 242 ± 6 |
| 100 | 2183 ± 58 | 1920 ± 51 | 269 ± 6 |

In addition to Table 2, Table 4 shows the average values of various leaching tests carried out in test tubes containing different families of conformed ceramics: in this case, for potential soil-contaminating chemical elements (arsenic, lead, and barium). Therefore, these results, which are statistically acceptable for having a reduced standard deviation for each family, were evaluated together with previous physical properties to obtain a high-quality ceramic material.

### *3.3. Data Mining and Fuzzy Logic*

Ceramics that conform with contaminated soil and clay must be potentially commercialized in a real scenario; consequently, these materials must have adequate physical properties and exhibit the leaching of polluting elements following regulations. There is also a fundamental property required for any ceramic material: that is, compression strength. For this reason, this property was taken in the fuzzy logic analysis as the consequent; this means that as a parameter influenced by the values of the rest of the variables within its range, depending on these antecedents, the consequent takes maximum, low, or intermediate values.

Therefore, the fuzzy logic methodology allows obtaining a series of cause–effect relationships between the evaluated physical properties and the compressive strength. As a novelty in this research study, this provides a combined percentage range of contaminated soil and clay that shows the feasibility of obtaining ceramic materials with sufficient resistance and, in addition, allows the determination of values that would be obtained in the rest of the variables. The results of the study with fuzzy logic are detailed in Figure 4, taking all previously evaluated physical properties as antecedents and compressive strength as a consequent.

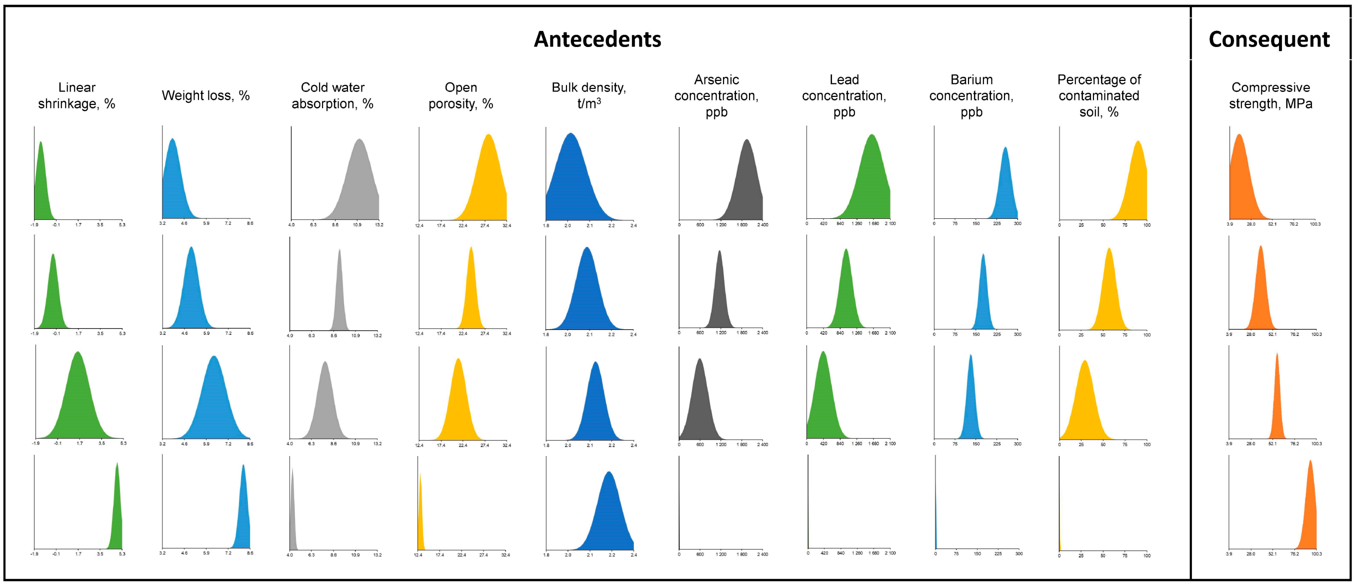


**Figure 4.** Fuzzy rules of the relation between physical properties (antecedents) and the compressive strength (consequent) of the ceramic material formed by clay and contaminated soil.

Figure 4, as mentioned, establishes the cause–effect relationships between the antecedents and the assumed consequent. Therefore, in a quick and very visual way, the existing relationships between variables can be observed, as well as the percentage of contaminated soil that must be added to obtain greater or lesser resistance. However, to understand Figure 4, a series of clarifications must be made, which are detailed below, and they reflect the depth of the study and the goodness of the results:

- Various physical properties of the conformed ceramics, as well as the concentration percentage of soil-contaminating chemical elements in the ceramic leachate, appear in the columns as antecedents.
- In each variable rule, there are numbers that correspond to the maximum and minimum value each variable can take in the tests. This range, which is essential for the analysis of results, is called the universe of discourse.
- Compressive strength appears separately and also in a column as the consequent established in the fuzzy logic study.
- With respect to compressive strength, the consequent shows different rules in the columns corresponding to various ranges of values. More specifically, and from top to bottom, the upper rule shows the range of values with respect to extremely low compressive strength, medium–low, medium–high, and, finally, extremely high compressive strength values.
- Therefore, with a simple glance at the rows, it is possible to determine the range of values that the antecedents must take to obtain high, low, or medium values of the consequent.

Based on what has been said and after observing Figure 4, it can be stated that for compressive strength to obtain extremely low to medium–low values, linear contraction and weight loss must also take extremely low to medium–low values. In addition, to achieve the aforementioned resistance, cold-water absorption, open porosity, and concentrations of arsenic and lead in the leachate must reflect extremely high to medium values. On the other hand, the barium concentration in the leachate and the percentage of contaminated soil in the ceramic must reflect extremely high to medium–high values in order to obtain low resistances with extreme–low results. The Gaussian bell curve obtained for the apparent density values is worth noting, and it is much wider and covers a greater range of values than, for example, linear contraction. This is due to the fact that in order to obtain extremely low to medium–low results with respect to resistance, density can take on several dispersed values, unlike linear contraction, because it is a variable with less dependence on resistance.

In turn, and completely contrary to the previous case, for clay, contaminated soil, and conformed ceramics to reflect medium–high and extremely high resistance results within the universe of discourse of this variable, it is unequivocally necessary that linear contraction and weight loss show values that fall between medium–high and extremely high. In addition, it is observed that cold-water absorption; open porosity; the percentage of contaminated soil in the ceramic; and the concentration of arsenic, lead, and barium in the leachate must take very specific values in the extremely low range to obtain higher compressive strength. The apparent density, as in the previous case, covers a range of important values within the universe of discourse, corroborating the previously mentioned hypothesis for this variable.

It should be noted that if Figure 4 is simplified to only show various variable trends of the function imposed on the antecedent, a greater or lesser dependency between variables is quickly observed, as well as the proportionality between them. This dependency is established by the slopes of the lines obtained by joining the gravity centers of various Gaussian bell curves, and they are more dependent on resistance than the variables that present a smaller slope. On the other hand, it is possible to observe the proportionality of the variables according to the direction of the subsequent lines; the variables are directly proportional if the two lines are approximately parallel and inversely proportional if they are convergent. Therefore, a simplified representation of Figure 4 is shown in Figure 5.

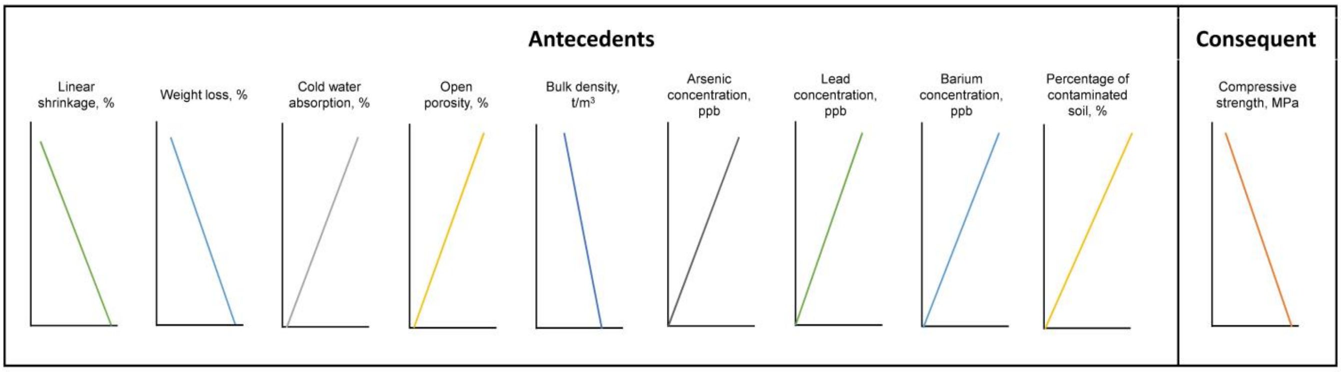


**Figure 5.** Simplification of the cause–effect relationships of antecedents and the consequent represented in Figure 4. This is based on the trend line of the mean values obtained from the tuples in Figure 4.

Figure 5 shows how the increase in the percentage of contaminated soil in the ceramic material produces a lower linear shrinkage and even linear expansion. This fact reflects a more open structure in the ceramic that is formed by contaminated soil, which consequently results in lower mechanical strength.

In addition, a higher percentage of contaminated soil in the ceramic results in lower weight loss. This is expected if one takes into account the loss on ignition of the contaminated soil and clay, which is much lower in soil than in clay. This lower weight loss, on the other hand, does not result in higher strength, but it is associated with lower strength, in turn resulting in lower weight loss.

In turn, the cold-water absorption of different ceramics with clay and contaminated soil is higher when the percentage of contaminated soil is higher; in turn, this higher percentage of contaminated soil implies a lower compressive strength of the ceramic. These results are logical if it is considered that a higher absorption of cold water implies a higher number of pores that are connected; in this case, the ceramic structure is much more open and develops lower strength. However, this property must be studied in detail for elements that are outdoors, since higher water absorption causes increased ceramic weight and, consequently, a higher load for the structure.

In accordance with the above, Figure 5 shows that higher porosity results due to the higher percentage of contaminated soil, and this leads to lower compressive strength with respect to the ceramic. The higher porosity can be of interest for ceramic applications in buildings, as it provides better thermal and acoustic insulation characteristics for different spaces.

It is worth noting that the relationship between bulk density and ceramic strength is lower. This can be affirmed by observing that the slope of the apparent density line shown in Figure 5 is much greater. Consequently, this means that density can present more diverse values to obtain the same strength result. This is expected if one looks at Figure 4, which shows how bulk density has much wider Gaussian bells and occupies a large part of the universe of discourse. However, it can be affirmed that a lower bulk density, with respect to ceramics, develops lower compressive strength, and this is conditioned by a higher percentage of contaminated soil.

Finally, it can be concluded that the higher strength of the ceramic is related to the lower concentration of chemical elements such as arsenic, lead, and barium in the filtrate. This result is predictable if one considers that higher compressive strength is associated with a lower percentage of contaminated soil, as the latter is responsible for the release of the aforementioned chemical elements. However, it is important to highlight that the filtered liquids of all families of ceramics made with clay and contaminated soil present concentrations of arsenic, lead, and barium below the limits established by the US-EPA for this type of material. Therefore, it can be stated that the ceramic matrix retains a large amount of toxic elements that are present in the soil and that the use of this procedure is feasible.

## 4. Conclusions

The study of materials, the development of the proposed methodology, and the analysis of the results with fuzzy logic techniques allowed justifying whether the objective of the present research has been achieved. This objective consists of the reuse of contaminated soils in new ceramic materials; on the one hand, this is carried out in order to reduce the environmental cost involved in the manufacture of ceramics, and on the other hand, it can prevent contaminated soil from producing environmental affections thanks to the retention of its contaminating elements in the ceramic matrix.

It has been shown that the use of contaminated soil in combination with clay for the creation of ceramic materials decreases the compressive strength of the element. This decrease in strength is directly related to the lower linear shrinkage and bulk density of the ceramic; moreover, the higher the porosity and cold-water absorption, the higher the percentage of contaminated soil in the ceramic.

However, the TCLP test has shown that the ceramic retained potentially contaminating chemicals in the contaminated soil; therefore, all families of ceramics were suitable for commercial use according to the detailed regulations.

Consequently, according to the partial conclusions reached, it is possible to use contaminated soils from the mining district of Linares, Spain, as raw materials for ceramic materials, and they can be used up to a high percentage. In addition, it was possible to prove the usefulness of fuzzy logic techniques within the circular mining concept, especially once the method provided results that were impossible to obtain using classical statistics. This new method established a combination field comprising clay and contaminated soil, and it is capable of producing marketable ceramic materials with different properties for various purposes; importantly, these results are predicable via fuzzy logic.

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