



Article An Innovative Experimental Petrographic Study of Concrete Produced by Animal Bones and Human Hair Fibers

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Abstract: The sustainable use of agricultural, industrial and human waste products as raw materials in the construction industry has been extensively investigated. This study aimed to conceive an innovative concrete composition mainly using recycled materials, which, as a result as waste from human activities and natural organic growth, such as animal bones and human hair, will be used in different mixtures in order to prepare concrete specimens. More specifically, the effect of these materials upon the final concrete strength was investigated, as well as how their petrographic characteristics may influence the durability of concrete specimens. Special emphasis was placed on the effect of the artificial increase in bone and hair microroughness and how these can improve the mechanical strength of the final product. The research results point towards the fact that the percentage of the replacement of natural aggregate rocks by animal bones tends to be enhanced by the increase in their microroughness using quartz primer. In addition, the use of bones with increased artificial microroughness and a certain percentage of human hair with increased microtopography seems to be the ideal mixture for the replacement of natural aggregates for the production of normal concrete.

Keywords: recycled materials; petrographic study; concrete; aggregates; human hair fibers; animal bones

1. Introduction

In recent years, the demand for modern constructions has vastly increased, making concrete the most important material. Regardless of the construction, concrete is used in every project, and as a result, it has become the second most consumable product after water. Along with this increased demand for concrete, there are two other important materials which constitute a major part of concrete: fine and coarse aggregates. There is a great demand for aggregates, mainly from the civil engineering industry for road and concrete constructions. On the other hand, the availability of fine and coarse aggregates constitute a serious problem, and for this reason, research efforts have been dedicated



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Copyright: © 2021 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 (https:// creativecommons.org/licenses/by/ 4.0/). towards developing waste management strategies in order to replace the traditional use of aggregates. The amount of construction waste is rapidly increasing year upon year in numerous countries such as India and China [1]. Natural resources are depleting worldwide, while at the same time, the generated wastes from different areas are substantially increasing. Currently, construction waste in many countries is mainly recycled in the form of roadbed bedding and recycled bricks. Sustainable development for construction involves the use of non-conventional, innovative materials as well as the use of waste materials in order to compensate for the lack of natural resources and involves finding alternative ways to conserve the environment [2]. In light of the EU Green Deal prospective and the recently issued New Circular Economy Action Plan, as well as to help achieve the United Nations Sustainable Development Goals (SDGs), it is necessary to promote the circularity of construction materials towards a carbon-neutral built environment by ensuring the appropriate use of resources with the least environmental impact.

Concrete, a mixture of cement, aggregates and water, constitutes the most used human-made material, with numerous civil engineering applications including those for the construction of roads, bridges and dams [3–6]. Any concrete structure can be described as a three-phase system composed of hardened cement paste, aggregate and the interface between aggregate particles and cement paste [7,8]. However, the rapid growth of the construction industry has led to environmental problems caused by excessive mining and by the extensive use of natural resources, including aggregate [9]. Approximately 275 million tons of new aggregate are annually extracted in the United Kingdom (UK), which is estimated to increase at a rate of 1% every year [10]. Concerns over the damage caused to the surrounding environment, the destruction of the ecological balance and the emission of greenhouse gases have become increasingly paramount [11]. The application of recycled aggregate has become a priority project in various countries and recycled aggregate concrete (RAC) is widely regarded as a new channel for the sustainable development of the construction industry [12–15]. In addition, improper waste recycling and the large number of industrial by-products have caused serious environmental pollution [16,17]. Therefore, there is a need to improve the reuse value of waste materials.

1.1. Use of Waste Materials as Concrete Aggregates

Various waste materials have been recycled and used as aggregates in concrete since the use of conventional aggregates is not environmentally friendly and contributes to the depletion of natural resources [18,19]. Various recycled waste materials have thus been used for the production of concrete such as: clay [20]; fly ash [21,22]; waste glass [23–27]; crushed bricks [27–30]; cathode ray tube glass waste [27,31,32]; crumb rubber waste [27]; ceramic and tile industry waste [27,33–38]; animal bones [39–41]; and plastic waste [27,36,37,42].

Several studies have been carried out to examine the impact of the use of glass waste on the behavior of the produced concrete, noticing that the content of glass waste influences the concrete's strength as well as its workability [23,38,43]. Research studies have been conducted on the use waste glass cullet in concretes, partially replacing natural aggregates, examining how the different proportions of waste glass cullet influence the produced concrete properties [23,24,27,43,44]. Moreover, the use of recycled ceramic tile waste as aggregate in concrete also contributes to relieving the industrial waste disposal problems and simultaneously to maintaining natural aggregate resources [45]. Several researchers have attempted the partial or total substitution of fine and coarse natural aggregates [27,38,46–48], investigating the produced concrete properties, while mainly taking advantage of the special properties of tile waste particles. Crushed asphalt, which is a typical by-product in the field of asphalt road rehabilitation, has also been used for the partial replacement of natural aggregates in concrete, contributing to the reduction in waste storage—which has dramatically increased in recent years [38,49].

Animal bones from goat or sheep are materials that are regarded as waste for the industry, and could be considered for partial usage in the construction industry as a substitute for either fine or coarse aggregates. In the present era, there are several animals that are

used to produce meat-based foods, including goats, sheep and cows. Large quantities from these waste bones are simply thrown in garbage bins and pits resulting in environmental degradation, and the pollution of land and water. Therefore, it is necessary to examine the potential for animal bones to be utilized beneficially in producing concrete, but most importantly to consider its use in offering high quality constructions. Therefore, an effort has been made to utilize these bones (crushed) to study the effect of animal bones on concrete. There are several scientists which have used animal bones in concrete specimens [39–41]. Bhat et al. [41] worked on machine-crushed animal bones as partial replacement of coarse aggregates in lightweight concrete. The following conclusions were drawn: Lightweight concrete using Crushed Animal Bone (CAB) aggregate can be achieved by replacing normal aggregate with approximately 50% or more CAB aggregate. The average unit weights corresponding to 50%, 75%, and 100% of CAB aggregate inclusion in concrete are 19.60 KN/m³, 17.65 KN/m³, and 16.55 KN/m³, respectively, for nominal concrete mix 1:1.5:3. Compressive strength of CAB concrete (lightweight) is low compared to normal concrete; however, it can be improved by using silica fume (SF). Besides achieving economy in construction, by reducing the weight of the structure, the catastrophic earthquake failures caused due to inertia forces (earthquake forces are proportional to the weight of the structure) that influence the structures can also be ultimately reduced.

Fapohunda et al. [50] investigated the suitability of crushed cow bone (CCB) as a partial replacement of fine aggregates for concrete production. The results showed that that there was a reduction in concrete workability, with an increase in the percent replacement of sand with CCB. The use of CCB also resulted in harsh mixes with attendant low slump. The density of the concrete specimens reduced as the percent increase in sand replacement with CCB increased. Using CCB as a partial replacement of sand can result in different types of concrete, based on the density attainable. The compressive strength of the specimens decreased with increase in the percent replacement of sand with crushed cow bone. The compressive strength of the specimens increased with curing ages. Replacement of sand with CCB up to 20% weight will result in compressive strength development that is not significantly different from those of the control samples. The use of CCB in the replacement of cement up to 20% weight in the production of concrete will have a positive impact on the environment, and encourage the use of bio-concrete in structural engineering.

1.2. Use of Hair Fibers in Concrete

Reinforced concrete has established itself as one of the major building materials for the improvement of the mechanical performance of the construction applications. One of the incentives for fiber reinforced concrete (FRC) is the possibility to reduce the amount of conventional reinforcement. The main differences between the conventional reinforced concrete and FRC concrete are the increased resistance and stiffness of the latter during the crack propagation stage, as well as that fact that it presents significantly smaller crack widths. It has been stated by [51] that after crack initiation, the crack propagates fast in the conventional concrete compared with the FRC. Moreover, the flexural stiffness is larger for the FRC member [51]. The reinforced concrete was patented by Joseph Monier in 1867. Since then, fibers have been utilized as support in concrete [52]. The great amount of human and animal hair fibers which are wasted annually has led scientists to use them in order to reinforce concretes, and, more specifically, hairs are utilized as a fiber strengthening material which impacts the compression, flexure and the cracking control. Fiber is frequently portrayed by a helpful parameter; the angle proportion which is the proportion of its length to the distance across. The use of fibers in concretes has well known advantages, as it helps in controlling the cracking, it reduces the porousness of concrete, it produces more impact resistance, and it also increases strength, ductility, shatter, abrasion resistance and delays cracking [52–60]. On the other hand, human hair presents properties such as unique chemical composition, high tensile strength, and high elasticity, which make it a useful material to be contained in concrete. However, a significant research gap concerning the effect of the artificial microroughness on the hair surface or in

general the further processing of hairs in order to significantly increase their properties has been observed.

This study aims to investigate the performance of an innovative concrete composition using mainly recycled materials arisen as wastes from human activities as well as from natural organic growth such as bones from different animals and human hair which will be used in different mixtures in order to prepare concrete specimens that are innovative and as environmentally friendly as possible. More specifically, the effect of these materials on the final concrete strength is investigated, as well as how the petrographic characteristics of the recycled materials may influence the durability of concrete. Special emphasis was placed on the effect of artificial microroughness of raw materials used as aggregates.

2. Materials and Methods

Animal bones from two different animals (goat and sheep) were used for the partial replacement of natural aggregates (limestones) in the produced concrete specimens. Additionally, human hair fibers have also been used to reinforce some of the concrete specimens, replacing commonly used glass fibers or synthetic fibers mainly of polypropylene. The proposed research study entitled "An innovative experimental petrographic study of concrete produced by animal bones and human hair fibers" (manuscript ID: sustainability-1204680) was checked by the Ethics Committee of University of Patras (Greece) regarding the ethical approach of using human and animal waste materials in order to produce an environmentally friendly reinforced concrete. This study focuses on the use of sustainable materials in construction applications, which are in accordance with the principles of circular economy, as the waste can be used as raw material for producing a new product (e.g., concrete). More specifically, waste animal bones were used to replace natural aggregates (e.g., limestone) and waste human hairs to replace other types of fibers (e.g., silicate, plastic) in order to produce an environmentally friendly reinforced concrete. The collected waste animal bones are derived from a butcher's shop (Patras, Greece) and the waste human hairs are derived from a barbershop (Aigio, Greece). Nevertheless, the energy required for treating animal bones (waste) as well as human hair is much less than that required for the re-extraction of natural aggregates, causing a much smaller environmental footprint.

Taking into account all the above, the competent committee based on the rules of the Declaration of Helsinki of 1975 (www.wma.net/what-we-do/medical-ethics/declaration-of-helsinki/, accessed on 15 April 2021), revised in 2013, decided with protocol number (ID: 7894) that the above study does not require any additional ethical approval. In addition, the same committee declared that no experiments were conducted using live animals and therefore this study does not fall within the limitations of the ARRIVE guidelines (www.nc3rs.org.uk/ARRIVE, accessed on 15 April 2021) for reporting experiments using live animals.

Normal Portland cement (CEM II 32.5N), which was conformed according to EN 197-1 [61], combined with the recycled materials (used as aggregates) was also used for the production of the concrete specimens. Potable tap water, free of impurities, such as salt, silt, clay, and organic matter, was used for mixing and curing the concrete. The pH value of the water was 7.0. In order to retain a consistent composition for all of the concrete specimens, we adopted the principle of maintaining the same volume of aggregate per 1 m³ of the mixture. The proportions and the volume content of each aggregate were calculated based on their different physical properties. The proportions of the concrete mixtures, by mass, were 1/6/0.63 ratio of cement/aggregate/water. The given ratio and proportion were chosen as they display better workability and compaction.

In this study, the animal bones, which were collected from disposals, were cleaned in order to partially separate them from the flesh, tissues and fats. In a next stage, they were heated in 92 °C in order to achieve the complete separation of any remaining flesh, tissue or fat that was still on the bone. Then, the dried bones were crushed in a jaw crusher in order to achieve the aggregate sizes needed for concrete aggregates (Figure 1a). The human hair samples, and more specifically men's hair that had not been dyed were collected from

the waste of a barbershop, and they were washed with acetone and air-dried before being included in the design concrete mix (Figure 1b). It should be mentioned that the human hair which was used and modified in this experimental study would not be necessary for any historical, medical or social application. Throughout the process of mixing the concrete components, the total volume of the human hair was placed periodically and in equal parts. Moreover, the mixing time was doubled in order to achieve the homogeneity in their distribution as well as in order to prevent the formation of agglomerates. The latter was incorporated into the volume of the produced concrete, where it was uniformly ensured that it was the first component added into the mixer when mixing the raw materials of the concrete. The concentration of human hair samples (length up to 15 mm) was 1.5% per volume, as it has been proven many times that this concentration is the optimum one [52]. Moreover, in the present study, we performed additional modification of human hair's microtopography where locally increasing its microtopography by using resinous quartz primer (Figure 1c).



Figure 1. Photos showing: (a) animal bones heated in 92 °C; (b) human hair fibers; (c) human hair fibers with quartz primer.

The animal bones which were used for the partial replacement of natural aggregates (limestones) in the produced concrete specimens were selected in the size classes of 4.45–9.5 and 9.5–19.1 mm, while the size class of 2.00–4.75 mm contained only limestones. An artificial increase in the microroughness of some of the used bones was carried out, since the microroughness of materials used as aggregates in concretes plays a critical role in their final strength, as it is responsible for the adequate bonding between the cement paste and the aggregate particle. In this study, the artificial increase in microroughness of the recycled materials was carried out by coating them with quartz primer three times in total, once every 24 h. The moisture content (w (%)) of the limestones used as aggregates in this paper was determined according to the [62] standard. Additionally, the moisture content of the animal bones used for the partial replacement of limestones was also determined before and after being coated with quartz primer.

Seventy-two normal concrete cube specimens (150×150 mm) from 18 different mixtures were made from the aggregate types (natural and not) according to ACI-211.1-91 [63] (Table 1). All of the parameters remained constant in all the concrete specimens. Workability of fresh concrete is measured in terms of slump loss by using standard slump cone according to specifications provided in the ASTM C143 [64]. After 24 h, the samples were removed from the mold and were cured in water for 28 days. Curing temperature was 20 ± 3 °C. These specimens were tested in a compression testing machine at an increasing rate of load of 140 kg/cm² per minute. The compressive strength of concrete is calculated by the division of the value of the load at the moment of failure over the area of the specimen. The compression test was elaborated according to BS EN 12390-3:2009 [65]. The loading rate according to the standards is 0.8 MPa/s and the test results should have an accuracy of 0.5 MPa. Furthermore, normal concrete specimens and human hair-reinforced concrete specimens of size 150 mm \times 150 mm \times 700 mm (according the mixtures presented in Table 1) were tested after 28 days of curing using a flexure testing machine in order to measure their flexural strength according to ASTM C78 [66]. The flexural strength was investigated in the laboratories of the Chemical Process & Energy Resources Institute, Centre

for Research & Technology Hellas (CERTH, Thermi, Thessaloniki, Greece). The quality cohesion between the cement paste and the aggregate particles (natural and recycled) in concretes were examined after the compressive strength test in samples oriented parallel to the force load. For this reason, two concrete particles and thin sections for each sample were studied in a polarizing microscope (Leica Microsystems, Leitz Wetzlar, Germany), according to ASTM C856–17 [67], as well as through scanning electron microscope (JEOL JSM-6300 SEM) equipped with energy dispersive spectrometer (EDS) and INCA software at the Laboratory of Electron Microscopy and Microanalysis, University of Patras. The operating conditions were accelerating voltage 25 kV and beam current 3.3 nA, with a 4 µm beam diameter. The bulk investigated rocks and animal bones were determined by powder X-ray diffraction (XRD), using a Bruker D8 Advance Diffractometer (Bruker, Billerica, MA, USA), with Ni-filtered CuK α radiation. The scanning area for bulk mineralogy of the samples covered the 2 θ interval 2–70°, with a scanning angle step size of 0.015° and a time step of 0.1 s. The mineral phases were determined using the DIFFRACplus EVA 12® software (Bruker-AXS, Billerica, MA, USA) based on the ICDD Powder Diffraction File of PDF-2 2006. Moreover, a 3D depiction of the petrographic characteristics of the concrete was carried out by the 3D Builder software using thin sections.

Table 1. Mixtures of recycled materials used as aggregates in concretes. (X, contained recycled material; -, not used recycled material; *, concrete specimens made with goat bones; **, concrete specimens made with sheep bones). The proportions of the concrete mixtures, by mass, were a 1/6/0.63 ratio of cement/aggregate/water.

Sample	Bones	Bones with Primer	Hair	Hair with Primer	Limestones
Size Class (mm)	4.45-9.5 & 9.5-19.1	4.45-9.5 & 9.5-19.1			2.00-4.75
S0A	-	-	-	-	Х
SOB	-	-	-	-	Х
S1A *	Х	-	-	-	Х
S1B *	Х	-	-	-	Х
S1C **	Х	-	-	-	Х
S1D **	Х	-	-	-	Х
S2A *	-	Х	Х	-	Х
S2B *	-	Х	Х	-	Х
S2C **	-	Х	Х	-	Х
S2D **	-	Х	Х	-	Х
S3A *	Х	-	-	Х	Х
S3B *	Х	-	-	Х	Х
S3C **	Х	-	-	Х	Х
S3D **	Х	-	-	Х	Х
S4A *	-	Х	-	Х	Х
S4B *	-	Х	-	Х	Х
S4C **	-	Х	-	Х	Х
S4D **	-	Х	-	Х	Х

3. Results and Discussion

Natural resources are depleting worldwide, when simultaneously the volume of different type of waste, organic (i.e., animal bones, human and animal hair) and inorganic (plastic, crumb rubber, industry tiles, glass, recycled aggregates), are substantially increasing and therefore the sustainable development for construction involves the use of non-conventional, innovative materials and waste materials in order to contribute to the reduction in the future extraction of mineral raw materials [2], as this growth is also associated with escalating impacts on the environment and society. This study was designed following the modern trends of the present day regarding the disposal of waste materials and the need to reduce the use of natural aggregates in construction applications, as their use leads to numerous environmental impacts, as has already been mentioned. Additionally, the cement production and aggregate extraction and processing may lead to a loss of arable/forest land, coupled with the loss of biodiversity, waste generation,

and resource depletion. Furthermore, quarrying, construction, and repair activities may negatively affect society, due to the noise and air pollution that arise during blasting at quarry/construction sites, transportation of materials, and repair activities which also lead to user inconveniences. It should also be noted that cement constitutes an energy-intensive component and accounts for 5–8% of global anthropogenic CO₂ emissions, as well as significant levels of SOx, NOx, particulate matter, and other pollutants. The use of waste materials contributes to the reduction in the transport cost of natural aggregates, as well as to the reduction in the energy needed for the excavation of natural ones. At the same time, the environment is protected according to the EU Green Deal prospective, and the issued New Circular Economy Action Plan, as a byproduct, is converted to the raw material for the production of a new product. According to these principles, several scientists have partially replaced natural aggregates with animal bones [40,41], while others [52,53,55,56] have added human hair fibers to reinforced concrete specimens, ensuring the appropriate use of resources with the least environmental impact.

3.1. Aggregate Properties

The results of the moisture content (w (%)) of the tested limestones which are used as aggregates in concrete specimens produced during this paper are 0.40% and 0.60% in S0A and S0B samples, respectively. As shown in Table 2, where the values of the moisture content of the used aggregate materials (natural and recycled) are presented, the obtained values of carbonate rocks (limestones) are significantly lower than those obtained for the recycled aggregates (animal bones). However, there is an evident difference in the values of moisture content in the samples that were coated with quartz primer. This is attributed to the special characteristics of the resinous primer, which seems to be able to penetrate the porosity of the bone, significantly reducing its ability to retain water in the structure. Another characteristic seems to be the reduction in the moisture content before and after the resinous quartz coating of bones (S1A–S4D), as the determined moisture content (w) of the used animal bones was 4.20% and 3.60%, respectively, before and after they were coated with quartz primer. This significant difference in the percentage of moisture content may contribute significantly to the degree of bonding of these fragments (recycled aggregates) with the cement paste, and several researchers have come to similar conclusions [38]. However, the type of bone used (goat or sheep) does not appear to significantly affect the bone's ability to adsorb water in its structure, as shown in Table 2. This fact indicates that the animal types used have a weaker effect than those which are coated with quartz primer on their ability to adsorb water to their structure.

The carbonate aggregates used as components of the produced concrete specimens do not present many cracks or impurities in their structure, as can be seen from the petrographic study. More specifically, limestones used display a micritic texture with numerous fossils and veinlets of microcrystalline calcite (Figure 2a). Likewise, the petrographic study also shows that bones are characterized by a healthy structure and the absence of constituent changes in growth from the inside to the outside of the bone, while significant mechanical fractures in their structure are missing (Figure 2b,c). According to the X-ray diffraction analyses, the studied limestones contain calcite (Figure 3a) and animal bones contain mainly calcite and apatite (Figure 3b). All the studied recycled raw materials are able to withstand the mechanical stresses within the concrete structure in terms of their microscopic characteristics; either they have a layered (Figure 2b) structure or a granular structure (Figure 2c). A particularly important parameter of the microscopic study of bones is the fact that the microstructure of bones has similar characteristics to those of rocks, and therefore their study will be based on similar criteria to those reported by many researchers who microscopically study the mechanical strength of rocks in concrete [38,68,69].

Sample	w (%) (Aggregate)	UCScon (MPa) (Mean Value)	Standard Deviation of UCScon (MPa)	FS (MPa)
S0A	0.40	30.0	0.9	3.51
S0B	0.60	30.0	0.9	3.51
S1A	4.20	23.0	2.0	3.40
S1B	4.10	24.0	1.9	3.41
S1C	4.08	24.0	1.9	3.41
S1D	4.08	24.0	1.8	3.42
S2A	4.00	25.0	1.6	3.45
S2B	3.97	27.0	1.5	3.46
S2C	3.98	26.0	1.5	3.46
S2D	3.97	27.0	1.4	3.45
S3A	3.95	27.0	1.5	3.49
S3B	3.80	28.0	1.5	3.48
S3C	3.82	27.0	1.5	3.49
S3D	3.76	28.0	1.4	3.48
S4A	3.67	29.0	1.2	3.51
S4B	3.67	29.0	1.2	3.51
S4C	3.65	29.0	1.2	3.52
S4D	3.60	30.0	1.0	3.52

Table 2. Moisture content (w (%)) of the used aggregates, mean value of the uniaxial compressive strength (UCScon (MPa)) test results of the 72 examined concrete specimens and Flexural strength (FS (MPa)) of the produced concrete specimens.



Figure 2. Photomicrographs of natural aggregates and animal bones used as aggregates in crossed nicols showing: (**a**) micritic limestone; (**b**) zoned bone (goat) with calcite; (**c**) amorphous bone (sheep) with cryptocrystalline calcite.

3.2. Concrete Properties

One of the most representative tests of concrete, and at the same time an indication of its behavior, is the workability. Studying the workability of the tested concrete, it was observed that those which contain no human hair display the greater slump loss. As human hair is added to the concrete specimens, a decrease in the workability is observed, as can be seen from Figure 4, which is presented below. This decrease in workability may be due to the lower amount of water available, as hair required some water to coat its surface. The



workability also decreases more in concrete that has human hair coated with quartz primer. These specimens present similar workability with the reference concrete specimens.

Figure 3. X-ray diffraction pattern of: (a) limestone; (b) goat's bone (Cc: calcite, Ap: apatite).



Workability

Figure 4. Chart of the workability of the concrete specimens (with the green color representing the reference concrete specimens).

Table 2 lists the results from the compressive strength test of the produced concrete specimens (UCS_{con}). The concrete strength ranged from 23.00–30.00 MPa after 28 days of curing. The validity of the compressive strength test results is indicated through the standard deviation parameter (Table 2). More specifically, the reference concretes (SOA and SOB) produced by carbonate rocks display the highest strength values of 30 MPa (Table 2). As for the specimens made by the animal bones which have partially been replaced the physical aggregates (limestones) in certain size classes, those which only contain animal bones present the lower strength values (23.00 and 24.00 MPa), while those made by animal bones coated with quartz primer and with human hair also coated with quartz primer are presented as more durable, with a present strength value of 29.00 MPa both for S4A and S4B samples.

These differences in the uniaxial compressive strength of the produced concrete specimens among the samples produced from animal bones (Table 2) show that modifying them (coating them with quartz primer), resulting in the artificial increase in their microroughness, significantly affects their final cohesion with the cement paste and hence increases the final concrete strength, enriching the international research gap. This is primarily due to the reduced ability of the coated aggregates to adsorb water to their structure, as shown in Table 1. This leaves the correct proportion of water free to be used by the cement during curing, thus producing an increased strength concrete as well as increasing the microroughness, which brings about an increase in the mechanical concrete strength. It is also obvious that even a small percentage of participation in human hair such as (S2A–S2D) within the concrete structure works positively on the mechanical strength of the concrete, as it seems to prevent the formation and spread of weakness points within its structure. Furthermore, the fact of the highly efficient use of human hair in the structure of concrete gives new perspectives in their use in combination with the bones.

Table 2 also lists the results from the Flexural strength of the tested concrete specimens where the values of the flexural strength seem to follow the trend of the values of the uniaxial concrete strength, indicating that the difference in the mixtures has a direct impact on all the mechanical properties of the concrete and not only on their uniaxial compressive strength (Figure 5).



Figure 5. Uniaxial compressive strength (UCScon (MPa)) vs. the flexural strength (Fs (MPa)) of the produced concrete specimens.

For example, samples S1A–S1D that do not contain any human hair fibers show neither increased strength nor flexural strength compared to samples containing human hair fibers covered with quartz resin primer, e.g., S2A–S2D, S3A–S3D and S4A–S4D. It also seems that the presence of human hair coated with quartz resin primer has a more positive effect on the flexural strength. Several researchers have found similar results regarding the positive effect of hair, such as Jameran et al. [70], Borkar et al. [58] and Alyousef [71].

3.3. Concrete Petrography

As a reference concrete, two concrete specimens were used, which contain fossiliferous limestones as coarse aggregates. In the following petrographic images (Figure 6a), it can be observed that the natural carbonate aggregates show good cohesion with the cement paste, a fact which is also proved by the absence of intense fractures. The good cohesion of limestones with the cement paste is also the main reason why these aggregates are widely used in the global production of concrete; however, these characteristics also lead to a constant demand for, and therefore increase in, mining activity for them, while simultaneously creating many environmental problems.

As seen in Figure 6a,b, limestone aggregate materials do not present any significant reaction zone between them and the cement paste, while they do not appear to have been broken transversely on their axis after the concrete has been crushed, and therefore the aggregates are considered ideal for concrete production in the present study.

As for the specimens made by animal bones, the presence of fractures in the bones, as well as between the bones and the cement paste, is observed (Figure 7a,b). Moreover, samples S1A–S1D are characterized by low microroughness and by unsatisfactory bonding with the cement paste, something that is indicated through the zone over the grains which acts as detachment zone for aggregates during the load. Furthermore, a possible factor that may have significantly affected this unsatisfactory cohesion is the increased ability of bones to absorb moisture in their structure, resulting in them using the water needed by

the cement for its complete hydration process. In sample S2A–S2D, where bones, which are used for the partial replacement of limestone aggregates, are coated with quartz primer, they present better cohesion with the cement paste (Figure 7c,d) than the other concretes produced by uncoated aggregate materials. As seen in the micro scale where the failures occur, the better bonding between the primer-coated aggregates is probably due to the increased roughness provided by the primer of high microtopography.



Figure 6. (a) Photomicrograph (sample SOA) and (b) secondary electron image (sample SOB) showing reference tested concretes with good cohesion between limestone aggregate grains and cement paste.



Figure 7. Photomicrographs of representative tested concretes showing: (**a**) animal bone aggregates (parallel nicols, sample S1A); (**b**) animal bone aggregates (sample S1B); (**c**) animal bone aggregates coating with quartz primer and human hair fibers (crossed nicols, sample S2A). Back scattered electron images showing microstructures at the interface between cement paste; (**d**) human hair fiber (sample S2B).

An additional reason for the better bonding is the significant reduction in the porosity of the bones due to their application with the resin primer which seems to have acted as a sealant in the pore channels, significantly preventing the adsorption of water where it is there primarily in the mixture, which results in smooth curing of the cement through hydration reactions and therefore increases their mechanical strength. The increase in the microroughness seems to create safe-toothed formations capable of adhering the cement paste to their structure in a satisfactory way, evenly distributing the loading tendencies of the applied forces. The type of bones seems to affect, to a smaller degree, the final concrete

strength against the increased influence where the degree of microroughness is shown to exert. However, although research studies have dealt with the effect of bone type, it seems that less attention has been paid to the modification of their microtopography as a major influencing factor.

These differences are mainly due to their differing ability to absorb water in their structure and, to a lesser extent, to their particular mechanical and microscopic characteristics, as shown in the diagram (Figure 8). From this correlation diagram, where the relationship between the bones' moisture content and the final concrete strength is described, samples SOA and SOB which contain only natural aggregates (limestones) have been deducted, as the participation of natural aggregates in all the tested recycled concretes remain stable (Figure 8). To these samples, human hair was also added, and they display partial cohesion with the cement paste, as it is shown in Figure 7d below, where one part of the human hair has been stuck with the cement paste, while the other part has been detached from the cement paste after the concrete compressive strength test.



Figure 8. Moisture content (w (%)) of animal bones vs. the uniaxial compressive strength (UCScon (MPa)) of the produced concrete specimens.

Scientists have devoted efforts to finding a use for materials such as animal hair fibers, human hair fibers, glass and steel fibers to reinforce concrete mechanical strength when the crack formation and crack propagation is reducing [52]. In this study, the addition of human hair in the produced concrete indeed seems to have helped in the prevention of crack formation, because of the presence of internal stresses due to concrete shrinkage and to concrete contractions/expansions due to temperature changes during the maturation of the concrete. Furthermore, the addition of human hairs uniformly distributed within the concrete structure appears to have significantly increased the tensile strength of the concrete, as it may have functioned as a kind of three-dimensional reinforcement capable of delivering the same physical and mechanical properties throughout its area. Moreover, the size of the fiber (human hair) used in its specific form was also a very important parameter, as no pelleting points were found during mixing with the concrete. The way in which human hair (fibers) were incorporated into the volume of the concrete, which was carried out evenly in the mixer, also benefited from the above-mentioned fact, as well as the fact that human hair was the first material which was added when mixing the raw materials of concrete.

Furthermore, human hair presents low microtopography and acts as an invisible retaining mesh of its structure. The next samples, S3A-S3D result from the partial replacement of natural aggregates (limestones) with animal bones. To this mixture, human hair coated with quartz primer was also added. In these concrete specimens, good cohesion among human hair fibers, bones, physical aggregates and cement paste is indicated in Figure 9a,b as well as presented numerically in Table 2. Furthermore, human hair coated with quartz primer present medium to high microroughness and seems to have positively worked in increasing the mechanical strength of the final product, as—apart from the three-dimensional internal reinforcement created by the modified human hairs-it seems to have helped significantly in preventing cracks which automatically leads to reduced permeability of concrete and thus prevents agglomeration (Figure 9b). The prevention of microcrack growth appears to be precisely due to the existence of quartz grains, as within them the growth of any microcracks seems to stop, acting as a kind of mechanical barrier. Minimizing the occurrence of microcracks also has a direct effect on increasing the mechanical strength of the material (concrete) over time. In the last mixture of recycled and physical aggregates used in the produced concrete specimens (S4A–S4D), as the animal bones as the human hair used have been coated with quartz primer and hence they display good cohesion with the cement paste indicated by the absence of fractures between these components (Figure 9c,d). When studying the microtopography of different concrete specimens in the Scanning Electron Microscope, it was observed that samples S4A-S4D presented the highest microtopography among all the other specimens. Samples displaying this increased microroughness in both bone and human hair are the best compared to other mixtures regardless of the type of bone because of this artificial increase in the microtopography of bones and hair which, in these mixtures, is found to prevent the widening of cracks due to the increased cohesion and they act as bridges by transmitting tensile stresses from surface to surface of the crack. Furthermore, the significantly reduced detection in microcracks in these mixtures compared to the rest indicates that the use of these raw materials of high microtopography (aggregates and bristles) improves the ability of the concrete to undertake the surface cutting, which may be attributed to its different workability in contrast to other mixtures and therefore to its different compact capacity. In addition, in the aforementioned mixtures, a particularly satisfactory adhesion between the human hairs (fibers) with the cement paste was found in contrast to the human hair without microroughness, which indicates a different mechanism of failure with absorption of energy before the failure. More specifically, it seems that human hairs of low microtopography, during the loading, are moving towards a progressive detachment with slow transmission of cracks, which in the future may lead to the formation of numerous microcracks.

All the above are enhanced either through the processing of microscopic images after the 3D depiction or through the study of the transition zone between the aggregate and the cement paste as it is shown in Figure 10a–d. All bones seem to present satisfactory cohesion with the cement paste, which does not appear satisfactory enough as their microroughness increases. At this point, it is worth mentioning that the bones have similar microtopographical characteristics to those of other natural sedimentary aggregates, as has also been observed by the Petrounias et al. [72]. In addition, it is observed that although the hair has lower microroughness compared to that of the other components of the concrete, it seems that it gives the required cohesion.

To study the interface and the reaction zone between the aggregate and the cement paste, SEM-EDX spectra were obtained. The results of these analyses indicate that there is no alkali silica reaction zone, because a gel-type zone or one that is not rich in elements such as K, Al, Na and Si disappears (Figure 11). Petrounias et al. [73], when investigating the existence or lack thereof of the alkali silica reaction zone in concrete specimens produced by natural aggregate rocks using the same analytical techniques (SEM-EDX spectra), identified alkaline silica gels. This study was carried out using a quartz-rich resin primer capable to react alone with the cement alkalis and to form an alkali silica reaction; Petrounias



et al. [38] reached similar results where no alkaline silica reactions were detected in the use of recycled materials as aggregates coated with quartz primer.

Figure 9. Photomicrographs of representative tested concretes showing: (**a**) animal bone aggregates and human hair fiber coating with quartz primer (parallel nicols, sample S3A); (**b**) human hair fiber coating with quartz primer (sample S3B); (**c**) animal bone aggregates and human hair fibers coating with quartz primer, as well as limestone aggregates (crossed nicols, sample S4A). Back scattered electron images showing microstructures at the interface between cement paste; (**d**) animal bone aggregate and human hair fiber coating with quartz primer (sample S4B).



Figure 10. (a–d): 3D depiction of the investigated concrete specimens.



Figure 11. (**a**,**b**) Back scattered image of a concrete specimen; (**c**–**e**) SEM-EDX spectra from the interface of an aggregate particle with the cement paste from the spots 1, 2, 5 as shown in Figure 11b.

4. Conclusions

Natural resources are depleting worldwide, when simultaneously the volume of different type of wastes, organic (i.e., animal bones, human and animal hair) and inorganic (plastic, crumb rubber, industry tiles, glass, recycled aggregates), are substantially increasing, and therefore the sustainable development for construction involves the use of non-conventional, innovative materials and waste materials in order to contribute to reducing the future extraction of mineral raw materials [2]. Another advantage of their use is the reduction in the transport cost of natural aggregates, as well as the reduction in the energy needed for their excavation. At the same time, the environment is protected according to the EU Green Deal prospective and the issued New Circular Economy Action Plan, as a byproduct is converted to a raw material for the production of a new product. According to these principles, several scientists have partially replaced natural aggregates with animal bones [40,41], while others [52,53,55,56] have added human hair fibers to reinforced concrete specimens, ensuring appropriate use of resources with the least environmental impact. The present study is an original work and essentially introduces the international literature in a new research object where it concerns the artificial increase in microtopography in general of recycled materials, so that it is possible to significantly increase the rate of the replacement natural aggregates. However, the use of quartz primer as used in the present study as a microroughness enhancement material is proposed to be replaced in the future by other primers of different mineralogical composition, but of equally important microtopography and hardness as the presence of excess SiO₂ is likely to create a number of alkali-silica reactions in the produced concrete specimens.

The present study examines the mechanical behavior of various concrete specimens made by the partial replacement of natural aggregates (limestones) by animal bones, either coated with quartz primer or not. In some of the mixtures, human hair coated with quartz primer (or not) was also added. The mechanical behavior of the produced concrete specimens was investigated using several petrographic methods and the most remarkable conclusions of the present work can be summarized as follows:

- The partial replacement of natural aggregates by bones can be carried out for the production of normal concrete specimens.
- The presence of animal bones with the artificially increased microroughness, after they
 have been coated with quartz resin primer of strong adhesion, seems to contribute to
 the final strength of concrete specimens made by the partial replacement of natural
 aggregates with these modified bones.
- With the addition of 1.5% per volume of human hair of certain size, it is possible to increase the homogeneity of the heterogeneous concrete mix, and therefore its final strength.
- Including a specific proportion of modified human hair (of high microroughness) in the concrete structure seems to have a significant effect upon increasing the strength of the final product.
- From the examined mixtures, it appears that the ones that contained animal bones with
 artificially increased microroughness and human hair of increased microtopography
 are the most ideal for the replacement of natural aggregates in normal concretes.

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