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Utilization of Granite and Limestone Wastes as Aggregates in Concrete

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The feasibility study of utilizing granite and limestone waste materials as alternatives to aggregates in various grain sizes in concrete is carried out in this work.

The granite and limestone wastes are pelletized into different grain sizes and then dried for 48 hours in open air at room temperature to remove the excess water and enhance workability. Subsequently, the pellets are fired in a rotary kiln at a very high temperature, with a heating rate of 20°C per minute, for 2 hours at 1100°C.

The chemical and mineralogical composition of the waste materials is analyzed using techniques such as petrography, X-ray fluorescence (XRF), X-ray diffraction (XRD), scanning electron microscopy (SEM), and energy-dispersive X-ray spectroscopy (EDX). These analyses show the composition and structure of the first material which is granite with a high iron content (6.66%) is called "Ferriferous granite", the second material is granite with normal iron content (3.15%) is called "Normal granite" and the third material is "Limestone", and aid in the selection of suitable materials for concrete production.

In conclusion, this research aims to investigate the potential use of granite and limestone waste materials as replacements for traditional aggregates in concrete production. Through a series of shaping, burning, and mixing processes. The important results obtained is that as the percentage of granite waste increases, the pellets become more friable. Therefore, based on this research, we recommend mixing the granite and limestone to enhance the durability of the aggregates, enabling their utilization in the production of high-strength concrete.

Keywords: Concrete, Waste Materials, Limestone-Granite Aggregates, Engineering Parameters.

1. Introduction

Natural stone mining and processing units release a significant amount of non-biodegradable waste into the environment in various forms, leading to some environmental impacts and hazards over time (Kumar et al., 2018). It is critical to find environmentally friendly ways to dispose of waste or repurpose it for value-added production.

In Egypt, an estimated 100 million tons of waste have accumulated in quarry areas over the past 15 years, hindering sustainable development. Unmanaged waste disposal near marble and granite factories has led to pollution of the air, soil, and water in nearby industrial, agricultural, and residential areas (Ciccu et al., 2005 and Garas et al., 2014). The generation of large amounts of stone-cutting waste poses both environmental and economic challenges. Using stone waste dust (SWD) from the ornamental stone industry and industrial production can alleviate environmental burdens. Uncontrolled dumping of SWD has demonstrated detrimental effects on public health (Gowaid et al., 2011 and Al-Tersawy et al., 2023).

To address this major environmental issue, researchers in Egypt have made preliminary attempts

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to manage the substantial amount of waste in the industry. The best approach has been to utilize these wastes in the construction sector, thereby reducing environmental pollution and preserving natural stones such as granite and limestone. As a result, ongoing research focuses on substituting aggregates with waste materials (Lakhani et al., 2014).

Additionally, the construction industry's high demand for natural resources for material production has prompted research into utilizing stone industry-based waste and byproducts in mortar development (Gehlot and Shrivastava, 2023). This involves replacing natural aggregates or fine aggregate/natural sand in mortar production (Gehlot and Shrivastava, 2023). Stones, including granite, marble, sandstone, and limestone, are widely used in the construction industry due to their versatility, cost feasibility, and availability Almeida et al., 2007, Tarbay et al., 2019 and Gehlot and Shrivastava, 2022. The quantities of waste materials and by-products are increasing due to the growth of construction industries, population, urbanization, development activities, and changes in lifestyle.

Recycling limestone and granite wastes offers cost reduction and eco-friendly production, as these industries generate large amounts of waste (Awad et al., 2019). Recycled materials such as granite, marble, and limestone dust are commonly used as fillers in construction. They contribute to improved strength, durability, workability, and sustainability, making them ideal choices for concrete manufacturing (Vollpracht et al., 2016, Arel and Shaikh, 2019, Tunc, 2019 and Al-Tersawy et al., 2023). While landfilling is an alternative, it is not sustainable, making reuse a viable option in terms of economic and ecological feasibility (Chahour and Safi, 2020). While many studies have explored the use of waste materials as fillers in Hot Mix Asphalt (HMA), only a few focus on analytical methods for predicting pavement performance (Gowaid et al., 2011 and Hamza et al., 2011). These stone wastes pose significant environmental challenges and threats to modern civilization, including drainage blockages, air pollution, dust nuisance, decreased land porosity and percolation, vegetation obstruction, loss of land fertility, non-biodegradable hazards, and water contamination (Chouhan et al., 2019). However, one potential solution to mitigate the environmental issues caused by the depletion of sands originating from rivers or mining is to substitute them with quarry waste in construction materials (Omar et al., 2012). This approach can help address the environmental challenges associated with sand depletion.

Various operations such as quarrying, sawing, cutting, and polishing are employed to shape raw stones into different products, utilizing modern technological tools that have increased waste production (Gehlot and Shrivastava, 2022). There are two main types of stone waste, Quarry/cutting/sawing waste and Polishing waste. Quarry/cutting/sawing waste, generated during the processing of raw stone blocks to obtain slabs and tiles while Polishing waste, generated during further processing to achieve smooth, shiny surfaces and textures (Gehlot and Shrivastava, 2022).

2. Materials and Methods

Granite wastes and limestone wastes are generated as by-products during the production of aggregates through the crushing process. Furthermore, these waste materials are also obtained as by-products from the sawing, shaping, and polishing processes (Corinaldesi et al., 2010). The accumulation of these waste materials raises important environmental concerns (Ergün, 2011).
Three waste materials (ferriferous granite, normal granite and limestone) with different origins were collected. Material 1 consists of granite with a high iron content resulting from contamination during the cutting process, while Material 2 contained granite with a low iron percentage compared with material 1. Material 3 was composed of limestone. To facilitate further analysis, the samples were shaped into coarse (4-6 mm) and fine (2-4 mm) aggregates. However, even after thorough drying at room temperature, the aggregates remained fragile and lacked the necessary strength for further studies. Consequently, it was decided to subject the samples to a burning process at 1100°C for two hours after molding in 5x5x5 moulds to enhance their strength. Following molding and firing, the samples were crushed, ground, and reshaped into fine and coarse aggregates once again. Upon drying at room temperature, it was observed that materials 1 and 2 still exhibited fragility, while sample 3 displayed significant strength as shown in Fig. 1.

To overcome the fragility issue, a decision was made to create mixtures of materials 1 and 2 with Material 3 and subject them to the same burning process. These mixtures, labelled as Sample A (25% from material 1

+ 75% from material 3) and Sample B (25% from material 2 + 75% from material 3), were molded into cubes and fired at 1100°C for two hours. After the second round of molding and firing, the resulting aggregates exhibited increased strength for both Sample A and Sample B (Fig. 1). Recognizing the improved properties of the mixed samples, it was agreed to explore different combinations.

Six new mixtures were prepared, molded into cubes, and fired accordingly. The mixtures included Sample A1 (25% from material 1 + 75% from material 3), Sample A2 (75% from material 1 + 25% from material 3), Sample A3 (50% from material 1 + 50% from material 3), Sample B1 (25% from material 2 + 75% from material 3), Sample B2 (75% from material 2 + 25% from material 3), and Sample B3 (50% from material 2 + 50% from material 3). These newly formed samples underwent firing, fine grinding, and reshaping into fine and coarse aggregates suitable for use in concrete (Fig. 1).

It is worth noting that, despite the improvements, Sample A2 and Sample B2 still exhibited a high degree of fragility. This observation suggests that as the proportion of granite increased in the samples, their friability also increased. This information provides valuable insights for further optimization and utilization of the aggregates in concrete applications.

Aggregate grains were investigated for their chemical and mineral composition by using XRD, XRF, SEM techniques and Petrographic Studies, respectively. XRD analysis was performed using a Philips X-Ray Diffraction equipment model X′Pert PRO with Monochromator, Cu-radiation (ʎ=1.542Å), operating at 50 K.V. and 40 M.A., with a scanning speed of 0.02º/sec. The XRD analysis covered the range of reflection peaks between 2ɵ =2º and 60º, providing information on spacing (d, Å) and relative intensities (I/Io). The diffraction charts and relative intensities are obtained and compared with ICDD files. The mineral composition of the materials is confirmed by the chemical composition that explains the major oxides and trace elements contents by using X-ray fluorescence (XRF). XRF analysis was performed using a PW 2404 X-ray fluorescence equipment. For the SEM analysis, a SEM Model Quanta 250 FEG (Field Emission Gun) equipped with an EDX Unit (Energy Dispersive X-ray Analysis) was employed. The SEM analysis was conducted using an accelerating voltage of 30 K.V. and a magnification range of 14x up to 1,000,000x. The SEM technique allowed for the acquisition of images that complemented the information obtained from optical and electron microscopy images.

3. Results

Different tests were carried out on the studied materials before firing, after firing at 1100 ◦C for 2 hours, and after mixing. These tests include chemical and mineralogical studies such as X-ray fluorescence, X-ray diffraction analysis (XRD), scanning electron microscopy (SEM & EDX) and Petrographic Studies.

Three materials were studied petrographically two granitic materials and one limestone. The first granitic rocks were composed of fine to medium-grained quartz, and feldspars (albite and microcline) as the main constituents associated with a minor amount of biotite, hornblende and muscovite whereas iron oxides occurred as accessory minerals (Fig. 2, A). Carbonates, sericite, chlorite and clay minerals occurred in the groundmass as secondary minerals.

The second granitic materials are soft, friable and of buff colour and composed of fine to medium-grained phenocrysts of quartz, feldspars and a minor amount of hornblende scattered in the matrix. The matrix is composed of very fine-grained quartz, clay minerals and feldspars as the major constituents with minor amounts of hornblende, muscovite carbonates and traces of biotite, iron oxides and opaques. Iron oxides occur as fine-grained aggregates and also as staining minerals over some parts of the matrix (Fig. 2, B).

The third material of limestone rock is soft, friable and creamy coloured. Microscopically, the sample is very fine to fine-grained. It is composed of carbonate minerals (mainly calcite) and is an essential mineral constituent associated with trace amounts of clay minerals, microcrystalline quartz, iron oxides and opaques. Carbonates and clay minerals represent the matrix of the sample and occur as very fine-grained aggregates. Quartz presents very fine-grained microcrystalline aggregates embedded in the matrix of the sample. Iron oxides and opaques occur as rare, very fine-grained crystals scattered in the sample. Some microfossils and shell fragments are observed and filled with recrystallized carbonates (Fig. 2, C). These mineralogical compositions of the investigated materials labelled as 1, 2, 3, A, and B are confirmed by XRD which are shown in Figures 3, 4, 5 and 6.

Fig. 1: Preparation of waste aggregates.

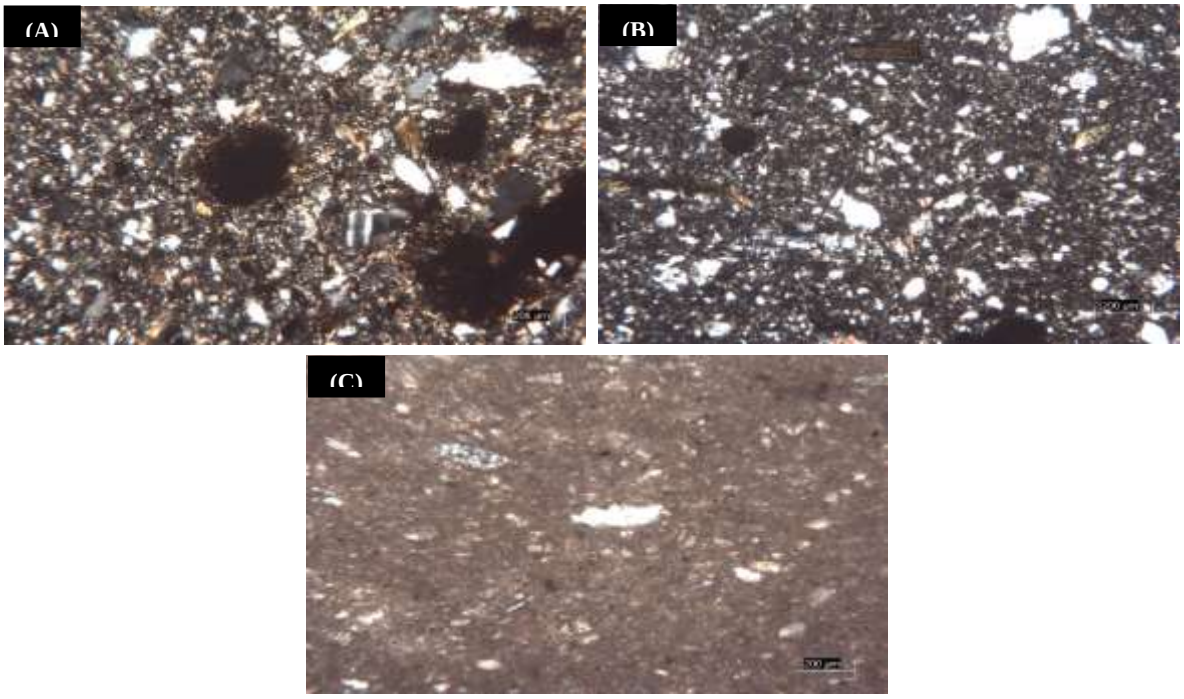

Fig. 2. Petrographic study of the three materials before firing, the first materials are granitic rocks (A), the second granitic materials (B) and the third materials are limestone rock (C).

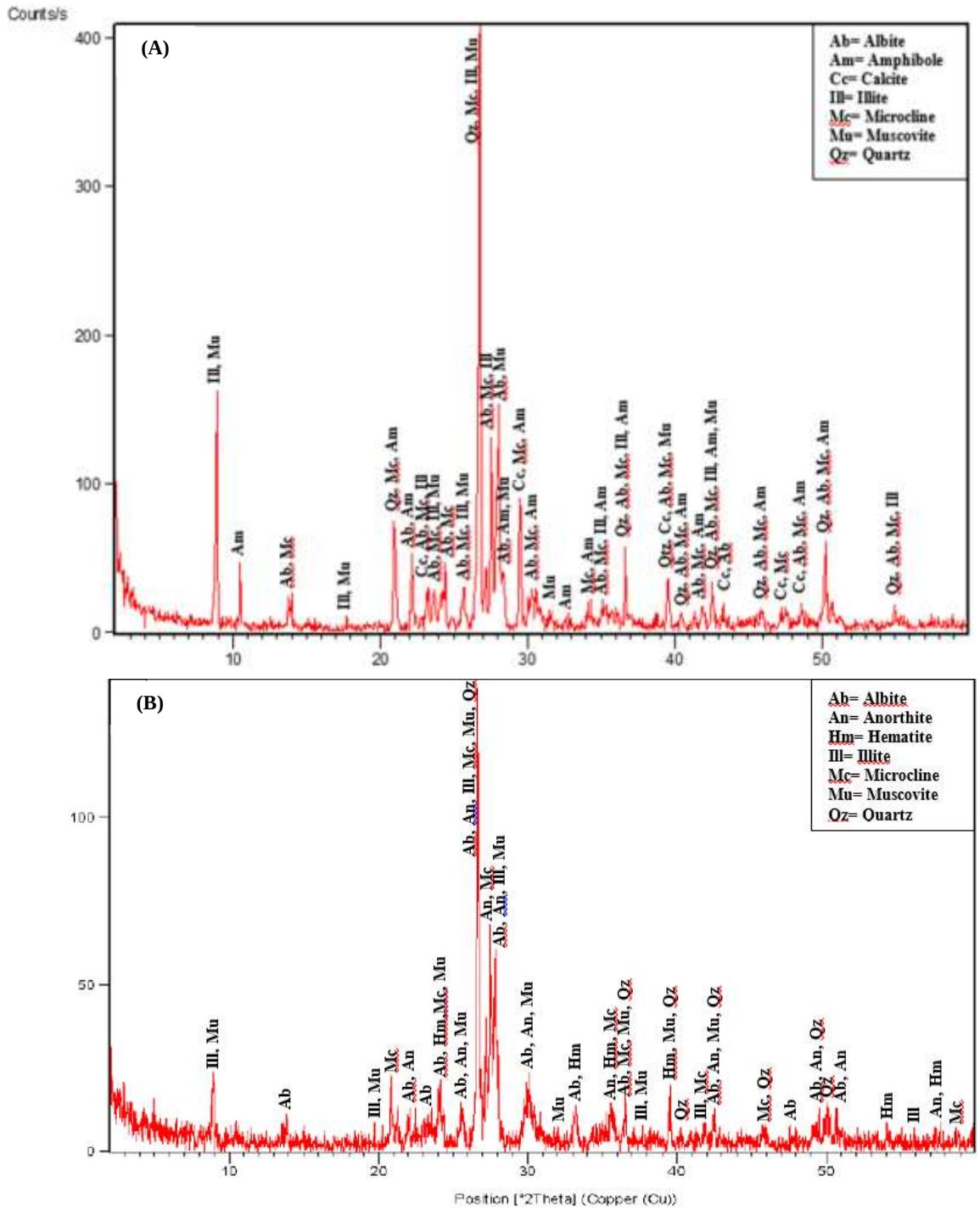

Fig. 3. XRD charts showing the mineral constituents of the studied ferriferous granite waste (material 1) before firing (A) and after firing (B).

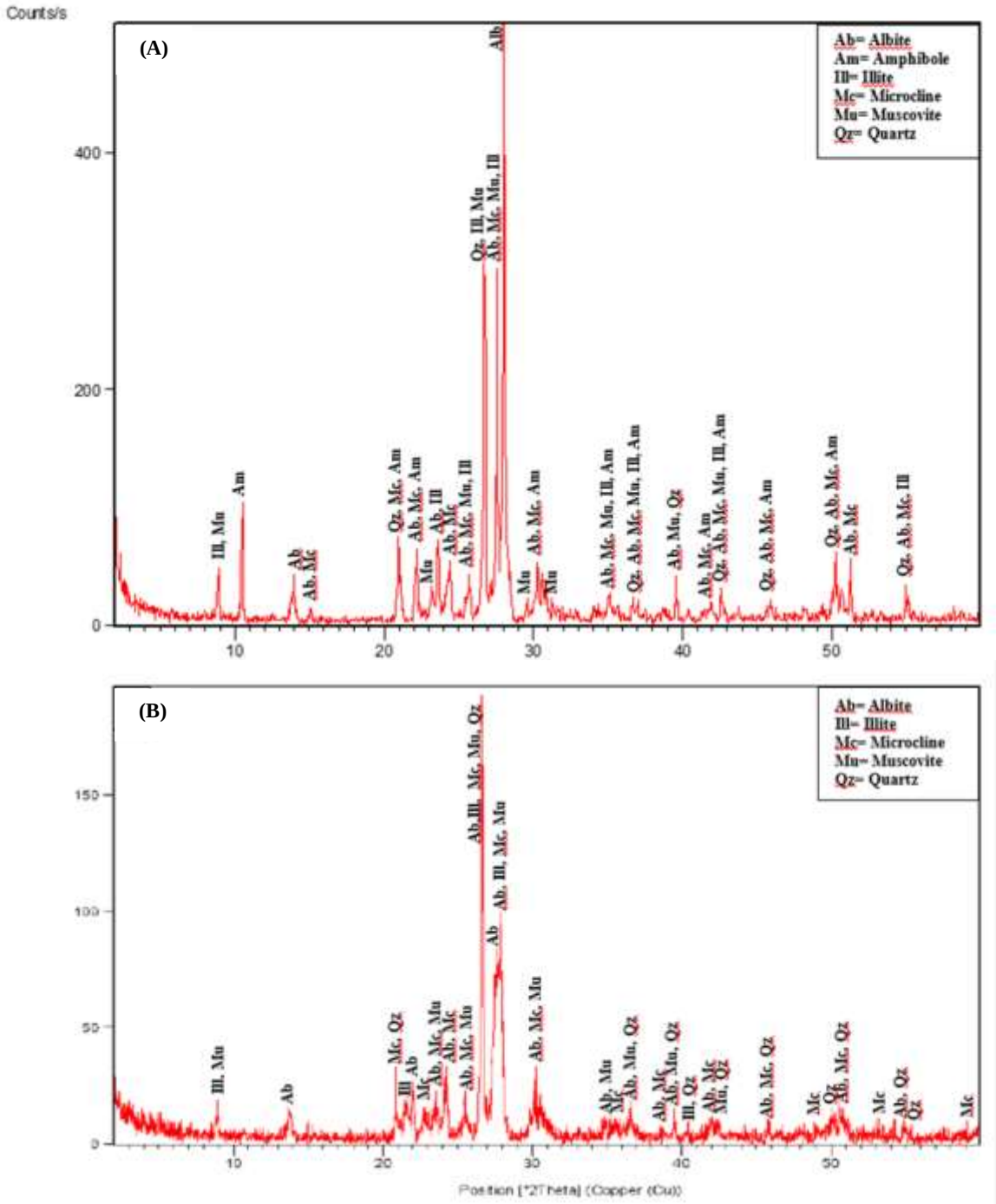

Fig. 4. XRD charts showing the mineral constituents of the studied normal granite waste (material 2) before firing (A) and after firing (B).

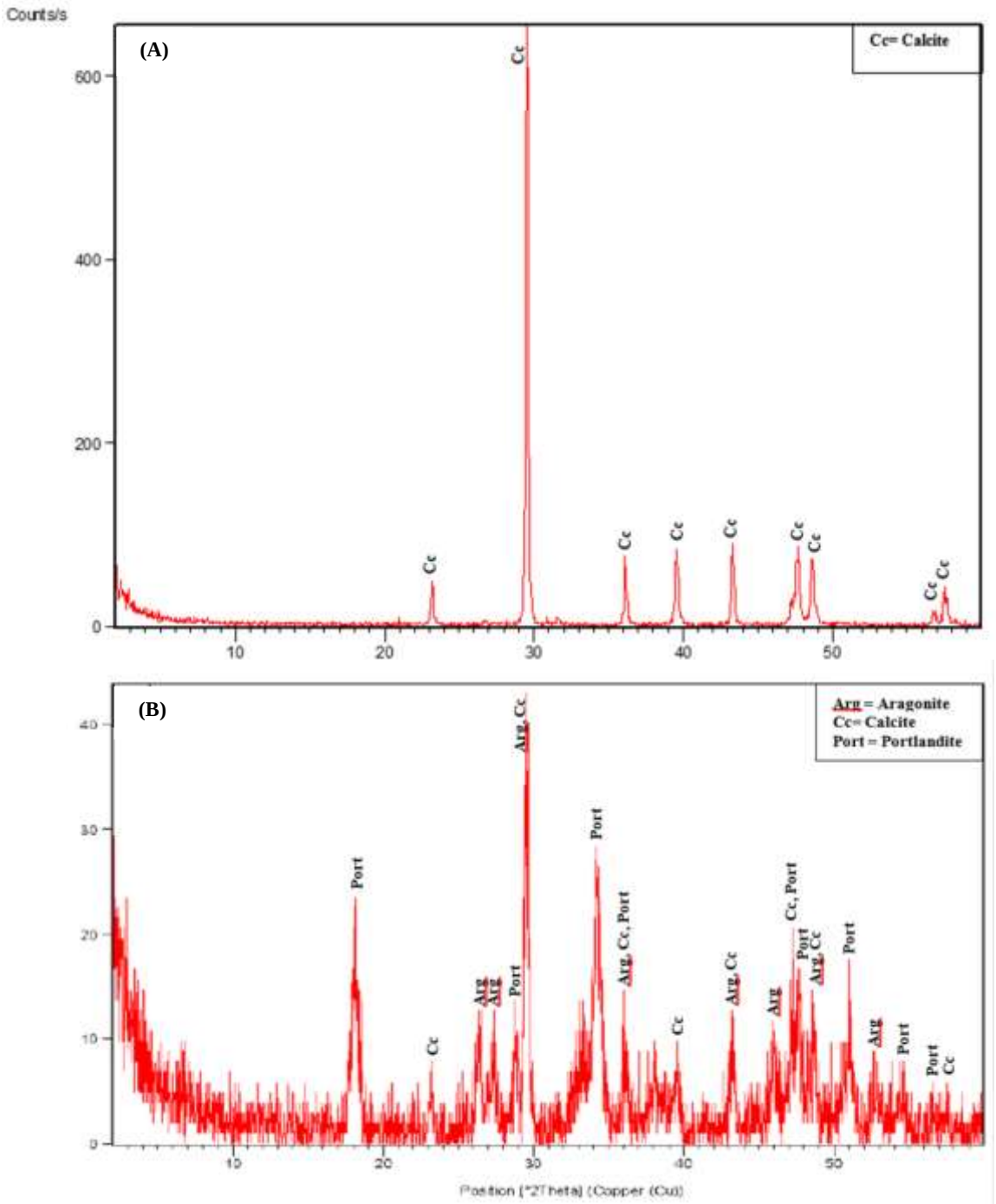

Fig. 5: XRD charts showing the mineral constituents of the studied limestone waste (material 3) before firing (A) and after firing (B).

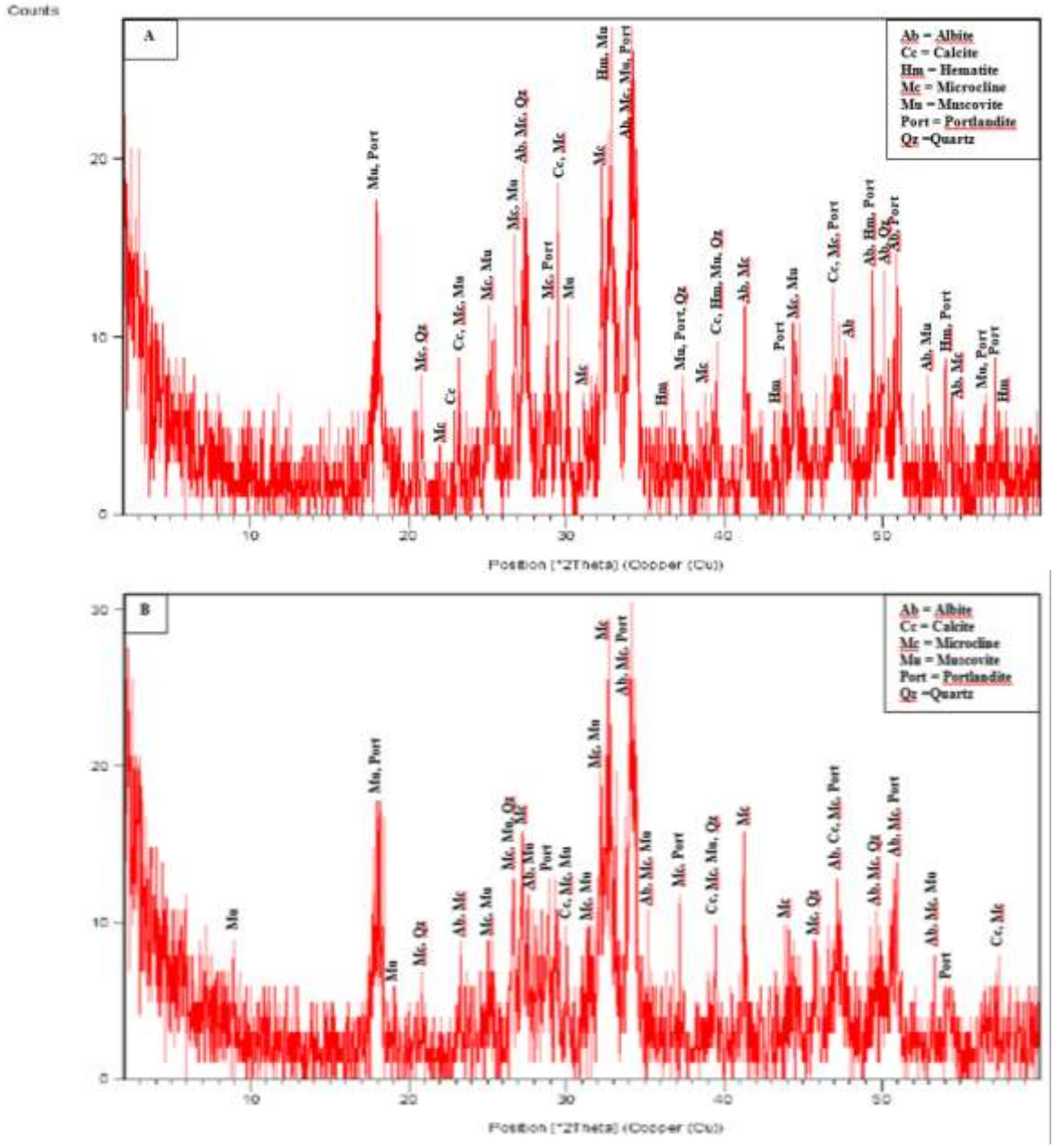

Fig. 6. XRD charts showing the mineral constituents of the studied samples A (A) and B (B) after firing.

This mineral composition of the materials is confirmed by the chemical composition by using XRF as shown in Table 1.

The chemical compositions of the material used in this study show the major oxide contents before and after firing to 1100 ◦C for two hours. All major oxide contents in materials 1 and 2 are slightly different whereas in materials 3 these oxides are changed especially calcium oxides. The calcium oxide in material 3 had a value of 55.58 before firing and changed to 69.34 after firing due to the loss of ignition decrease after firing (Table 1). The trace element composition of the studied materials 1, 2 and 3 before and after firing is presented in Table 2. It is noticed from the analysis shown in Table 2, that the high anomalous of Zr and Ba in materials 1 and 2, and the high anomalous of Sr in materials 3 are under investigation before and after firing.

The mineral composition of mixed samples composed from the three materials after firing (A and B) (Figure 6) is confirmed by the chemical composition (Tables 3 and 4). The chemical composition of sample A shows the major oxides contents of CaO is 70.93% and CO_2 is 16.43% and the low amount of SiO_2 is 7.38%, Fe_2O_3 is 2.09%, and MgO is 1.02% after firing (Table 3), whereas sample B contains the major oxides of CaO (73.41%) and CO_2 (12.27%) and low amount of SiO_2 (9.34%), Fe_2O_3 (1.28%), and MgO (1.14%) (Table 3). The trace element composition of the studied samples A and B after firing is presented in Table 4. It is noticed from the analysis shown in Table 4, that the high anomalous of Sr, Ba and Zr.

Table 1. Chemical Analysis (Major Oxides %) of the studied materials 1, 2, 3 before & after firing.

Oxide%	1	1	2	2	3	3
	Before Firing	After Firing	Before Firing	After Firing	Before Firing	After Firing
SiO_2	63.96	63.21	71.20	72.10	0.30	0.15
Al_2O_3	11.37	11.49	13.48	13.30	0.04	0.02
CaO	6.21	6.76	0.67	0.81	55.58	69.34
Na_2O	3.39	3.06	5.32	4.47	<0.01	<0.01
K_2O	4.16	4.40	4.15	4.56	0.01	<0.01
MgO	0.05	0.25	0.08	0.33	0.08	0.65
MnO	0.06	0.09	0.06	0.09	0.01	0.01
Fe_2O_3	6.66	10.07	3.15	3.71	0.07	0.08
TiO_2	0.38	0.32	0.36	0.34	0.01	<0.01
P_2O_5	0.01	0.04	0.01	0.02	<0.01	0.02
Cl	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
SO_3	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
L.O.I.	3.36	0.04	1.20	0.01	43.67	29.40

Table 2. Chemical Analysis (Trace elements ppm) of the studied materials 1, 2, 3 before & after firing.

Element (ppm)	1	1	2	2	3	3
	Before Firing	After Firing	Before Firing	After Firing	Before Firing	After Firing
V	35	27	18	19	7	12
Cr	150	234	<2	<2	30	9
Ni	19	45	14	16	6	5
Cu	17	42	83	66	10	9
Zn	89	68	115	97	18	15
Co	23	34	8	19	6	4
Ga	29	22	34	26	11	10
Rb	103	81	91	77	13	9
Sr	217	173	160	128	646	527
Y	50	38	50	41	16	12
Zr	399	310	435	350	71	55
Nb	36	26	53	43	14	10
Ba	904	776	700	616	<2	<2
La	112	102	115	101	12	<2
Yb	4	14	<2	5	<2	<2
Ta	2	<2	2	<2	3	<2
Pb	17	10	13	8	2	<2
Th	8	4	<2	5	20	16

Table 3. Chemical Analysis (Major Oxides %) of the mixed samples A & B after Firing.

Oxide %	A	B
SiO_2	7.38	9.34
Al_2O_3	1.07	1.34
CaO	70.93	73.41
Na_2O	0.01	<0.01
K_2O	0.59	0.67
MgO	1.02	1.14
MnO	0.05	0.05
Fe_2O_3	2.09	1.28
TiO_2	0.06	0.06
P_2O_5	0.05	0.03
Cl	<0.01	<0.01
SO_3	<0.01	<0.01
L.O.I.	16.43	12.27

Table 4. Chemical Analysis (Trace elements ppm) the mixed samples A & B after Firing.

Element (ppm)	A	B
V	19	15
Cr	66	6
Ni	13	10
Cu	20	36
Zn	35	57
Co	11	7
Ga	15	17
Rb	34	41
Sr	461	438
Y	22	27
Zr	156	202
Nb	18	28
Ba	212	180
La	32	31
Yb	<2	<2
Ta	2	2
Pb	4	3
Th	19	20

Scanning electron microscope (SEM) and energy-dispersive X-ray (EDX) analysis were conducted on materials 1, 2, and 3 before firing, revealing the presence of interfacial, interstitial spaces, pores and compacted surface. These findings explain the friable nature of these samples (Figure 7).

After firing, SEM and EDX analysis were performed on materials 1 and 2. The results showed the persistence of interfacial, interstitial spaces, and pores, which clarifies why these samples remained friable even after firing. In contrast, material 3 exhibited a compacted surface, indicating why this particular sample became significantly stronger after firing (Figure 8).

Upon mixing and firing to get samples A and B, SEM and EDX analyses were conducted, revealing a compacted surface. This finding explains why these samples became very strong after the mixing and firing processes (Figure 9).

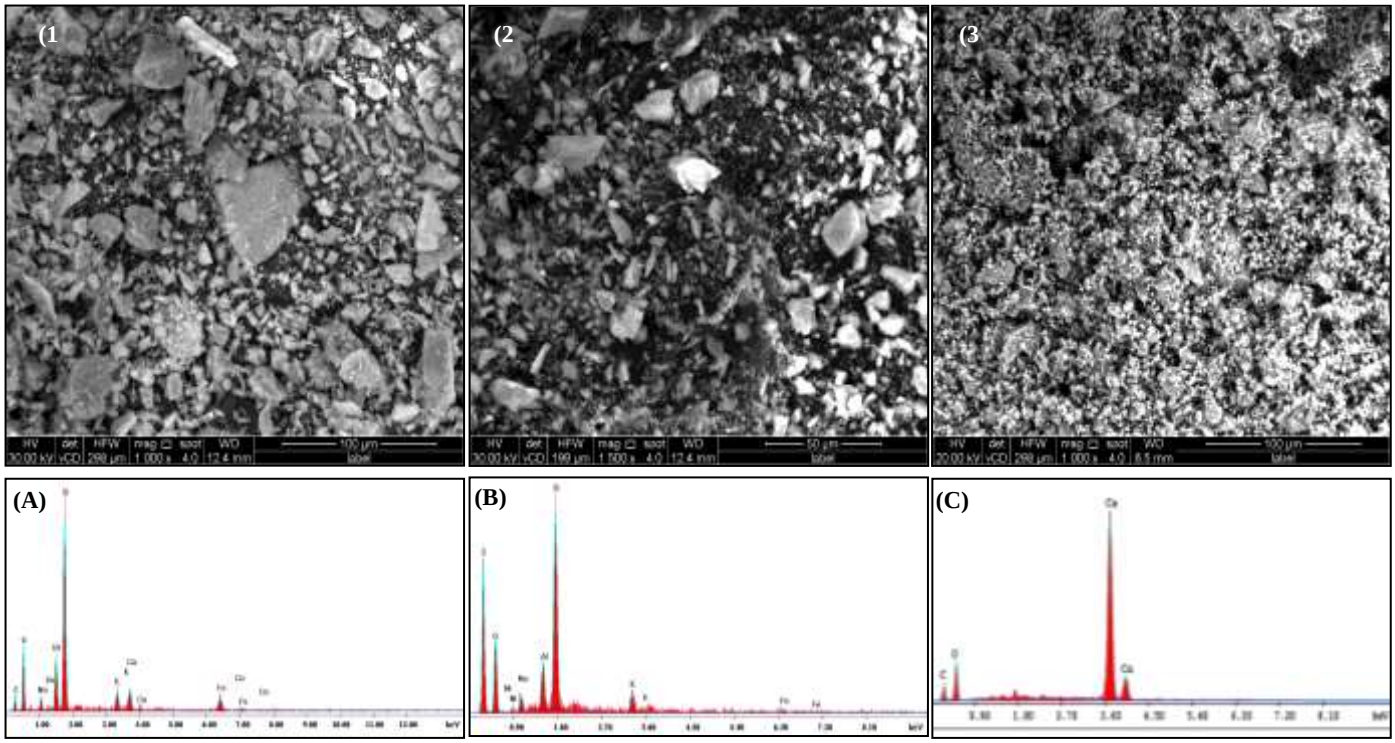

Fig. 7. SEM micrograph of the three studied materials 1, 2 and 3 before firing showing interfacial, interstial spaces and pores (1), (2) and (3) respectively. With bulk EDX of material 1 showing high silica with Al content and some iron (A), material 2 showing high silica with Al content (B) and material 3 showing high Ca content (C).

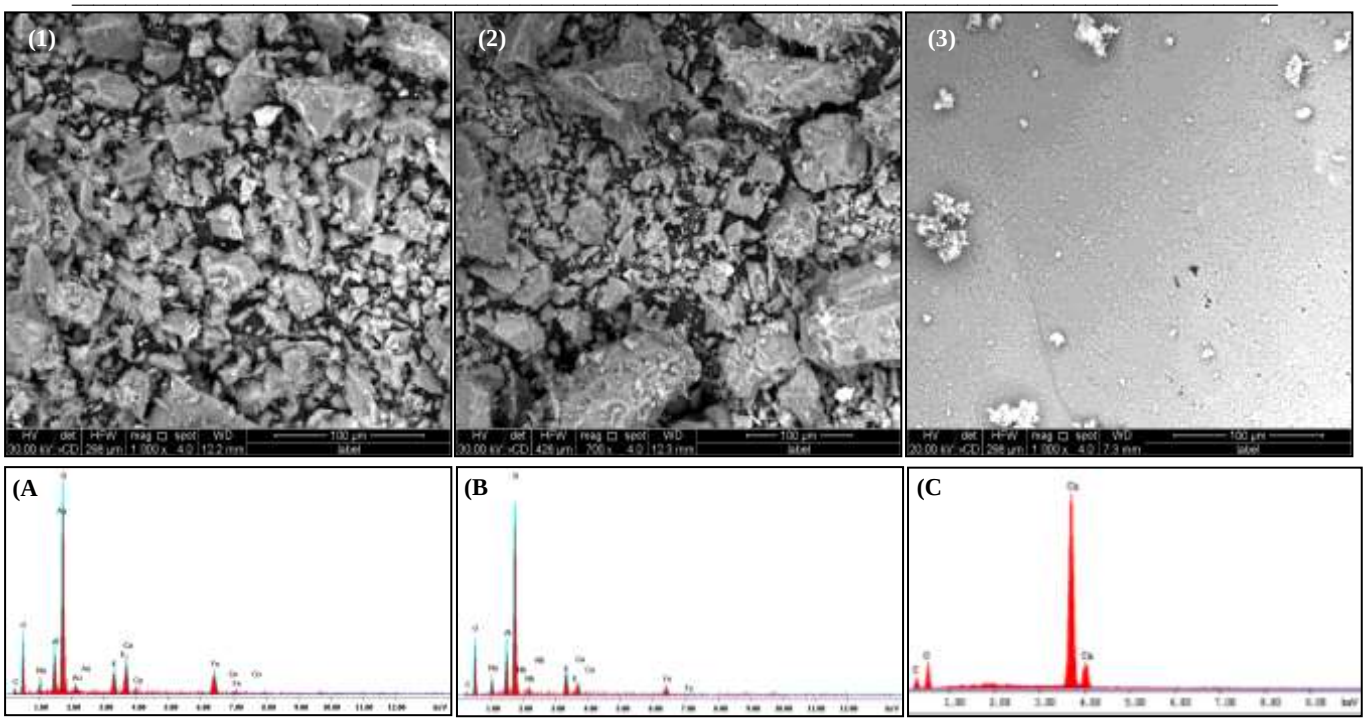

Fig. 8. SEM micrograph of the three studied materials after firing, materials 1 and 2 showing interfacial, interstial spaces and pores (1) and (2) respectively but material 3 showing compacted surface (3). With bulk EDX of material 1 showing high silica with Al content and some iron (A), material 2 showing high silica with Al content (B) and material 3 showing high Ca content (C).

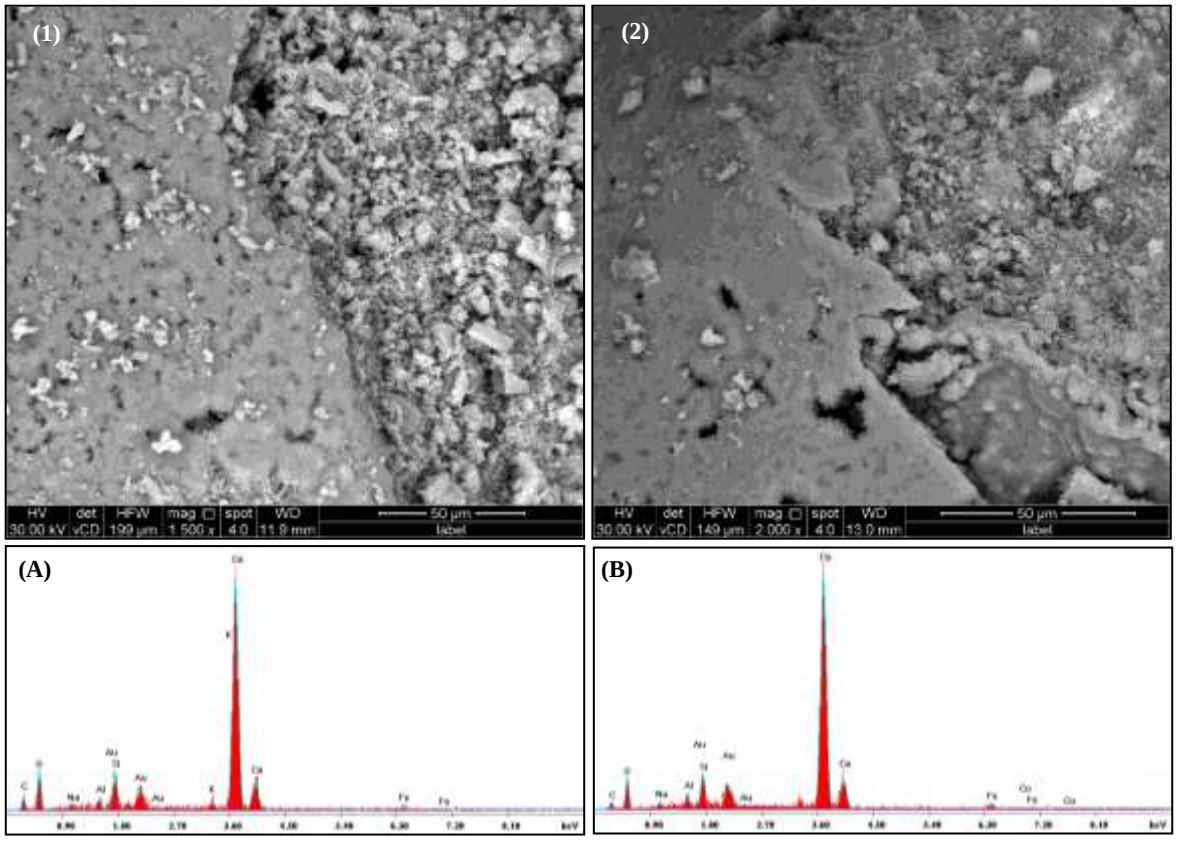

Fig. 9. SEM micrograph of the samples A and B after firing showing compacted surface (1) and (2) respectively. With bulk EDX of them showing high Ca with silica and Al content in both (A) and (B) respectively.

5. Conclusions

The primary objective of this study is to explore the possible utilization of granite and limestone waste materials as substitutes for conventional aggregates in the concrete production. This research involves a sequence of procedures, including shaping, burning, and mixing, in order to assess their viability, samples with improved strength were obtained. Granite wastes and limestone wastes are by-products generated during the production of aggregates and from the sawing, shaping, and polishing processes in the stone industry. The accumulation of these waste materials raises environmental concerns. To address the depletion of natural river and mining sands, substituting them with quarry waste in construction materials is a potential solution that can help mitigate environmental challenges.

To investigate the potential use of these waste materials as aggregates in concrete, samples were collected from different origins, including granite with high and low iron content and limestone. The samples were shaped into coarse and fine aggregates but remained fragile and unable to form rigid aggregates for further studies. To enhance their strength, the samples were subjected to a burning process at 1100°C for two hours. After moulding and firing, the samples were crushed, ground, and reshaped into fine and coarse aggregates.

Observations showed that materials 1 and 2 (granite with high and low iron content) remained fragile, while material 3 (limestone) exhibited significant strength. To improve the strength of materials 1 and 2, mixtures with material 3 were prepared and subjected to the same burning process. The mixed samples (A and B) exhibited significant strength after moulding and firing. Six new mixtures were prepared with different ratios of materials 1, 2, and 3, and they were fired, finely ground, and reshaped into fine and coarse aggregates for use in concrete.

Chemical and mineral compositions of the aggregate grains were investigated using XRF, XRD, Petrography and SEM techniques. XRF analysis provided information on the chemical composition, XRD analysis helped identify mineral phases, and petrography and SEM analysis allowed for detailed imaging and analysis of the samples.

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استخدام مخلفات الجرانيت والحجر الجيري كركام في الخرسانات

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نتيجة لوجود مخلفات ناتجة عن تقطيع صخور الجرانيت والحجر الجيري والتي تسبب أضراراً جسيمة بالصحة العامة وكذلك أضرار بالبيئة المحيطة، لذا كان من الضروري الاستخدام الأمثل لهذه المخلفات في الأغراض الصناعية، وقد تم استخدامها كركام في الخرسانات موضوع هذا البحث.

لقد تم اختيار الجرانيت والحجر الجيري نظراً لتواجدهم الواسع في مصر، واثناء استخدامهم كرخام واحجار زينة يصاحبهما كميات كبيرة من المخلفات التي تم توظيفها كركام في الخرسانات. وبناءاً عليه تم تجميع المواد المستخدمة وتم توصيفها معدنياً باستخدام جهاز حيود الأشعة السينية ودراسة تركيبها الكيميائي باستخدام جهاز الأشعة الفلوريسينية وايضاً تم وصف الشكل والتركيب الداخلي للعينات عن طريق الميكروسكوب الالكتروني الماسح.

تم تشكيل مخلفات الجرانيت والحجر الجيري على هيئة ركام بأحجام حبيبية مختلفة وحرق عند درجة حرارة ١١٠٠ درجة مئوية بمعدل تسخين يبلغ ٢٠ درجة مئوية في الدقيقة لمدة ساعتين وتم عمل خلطات من العينات اما منفردة او بنسب مختلفة بين نوعين مخلفات الصخور المستخدمة من الحجر الجيري والجرانيت وتم حرقها لزيادة متانتها وتم استنتاج ان كلما زادت نسبة الحجر الجيري عن الجرانيت في العينات كلما كان الركام المتكون أفضل للاستخدام في الخرسانات.