Investigation of Mechanical Properties and Microstructure of Construction- and Demolition-Waste-Based Geopolymers

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Abstract: Construction and demolition waste (CDW) is the third-most abundant waste generated annually in the countries of the European Union. One of the alternatives to the use of these wastes is geopolymeric materials. Partial replacement of commonly used raw materials for the production of these materials can help reduce the number of landfills and the consumption of natural resources. In this study, the authors partially replaced metakaolin and fly ash with clay bricks and concrete debris. The research method in article is connected with analysis of microstructures and the mechanical and physical properties of the geopolymers. The results obtained show the possibility of manufacturing useful construction materials based on industrial byproducts (fly ash) and CDW. Compressive strength and flexural strength were, for samples containing metakaolin, 20.1 MPa and 5.3 MPa, respectively. Geopolymers containing fly ash displayed 19.7 MPa of compressive strength and 3.0 MPa of flexural strength. The results for both synthesized materials give them perspectives for future applications in the construction industry.

Keywords: geopolymer composite; construction and demolition waste; circular economy

1. Introduction

The European Union has adopted two regulatory packages moving towards a circular economy. The first of these packages was introduced in 2015 with the announcement of the Circular Economy Action Plan and addressed five priority sectors where changes would accelerate the transition to a circular economy [1]. The changes and new regulations addressed plastics, food waste, critical raw materials, construction and demolition waste, biomass, and intermediates. In December 2019, the European Commission announced the European Green Deal, a development strategy to accelerate the transition to a circular economy [2]. In general, a circular economy is defined as an economy “where the value of products, materials and resources is maintained in the economy for as long as possible and the generation of waste is minimized” [1], which manifests itself as the efficient use of natural resources and the minimization or complete elimination of waste. To efficiently use materials, the key is to design a system in which a closed-loop process allows the minimization of material consumption while simultaneously allowing the economy to reuse or recycle said resources [3].

Construction is one of the industries that generates a significant amount of waste, accounting for 25–30% by weight of total industrial waste [4]. In countries of the European Union, the construction sector produces approximately 800 million tonnes of construction and demolition waste (CDW) annually [5]. This is one third of the total amount of waste produced each year [6]. Until now, sustainable disposal in the manufacturing process of structural and nonstructural concrete has been considered the best solution [7,8]. In addition, the increased demand for construction materials [8], the increased degree of industrialization [9], and the depletion of natural resources [4] force the further development
of materials based on alternative raw materials. The use of waste materials to produce construction materials is a desirable development direction [10], but further research is necessary to improve their properties.

A group of materials known for waste utilization is geopolymers. They are inorganic polymeric materials based on aluminosilicate precursors (such as fly ash, metakaolin, blast furnace slag, etc.) [11] activated by alkaline activators (mainly sodium, potassium) [12,13]. The final properties of the materials obtained depend on aluminosilicate precursors as well as the activator used, including their molar ratio [14,15]. Given their strength properties, which are comparable to those of conventional concrete, and much smaller carbon footprint, they are drawing considerable attention among researchers [16]. One of the directions of research on geopolymers is the search for alternative sources of aluminosilicate precursors. To advance the circular economy goals, materials with considerable potential are brick and concrete waste.

Şahin et al. in 2021 [17] compiled most of the research performed on geopolymers based on construction and demolition waste (CDW); the conclusion was the possibility of using the materials in the production of geopolymers, although more research is needed to develop knowledge about this group of materials [17,18].

Aldemir et al. [19] conducted studies of geopolymer composites based on demolition construction waste. The geopolymer composites included clay brick, tile, hollow brick, concrete waste (rubble), and glass. As an alkaline activator for the geopolymerization of these materials, 8 M sodium hydroxide dissolved in water was used, to which other activators—calcium hydroxide and sodium silicate—were added after 6 h. Two types of geopolymer concrete were produced, including: (1) NGC—tile, red clay brick, hollow brick, glass, concrete waste, slag, fly ash, and natural aggregate and (2) NGC-R—tile, red clay brick, hollow brick, glass, concrete waste, slag, fly ash, and recycled aggregate. All CDW materials were crushed and milled. The compressive strength tests of the samples showed values of 37.5 MPa and 36.6 MPa, respectively. In turn, the tensile strength values were NGC—2.56 MPa and NGC-R—2.37 MPa [19].

Komnitsas et al. [20] investigated the compressive strength of concrete-based geopolymers, bricks, and demolition tiles according to the grain size of the raw materials. Compressive strength was tested 7 days after samples were produced. The demolition materials were activated with 8 and 10 M sodium hydroxide solution with sodium silicate and water. The hardening temperature was 80–100 °C. The best results were obtained for geopolymers based on ceramic tiles at 57.8 MPa and bricks at 49.5 MPa. In turn, geopolymerized concrete achieved 13 MPa after 7 days of aging. The study also showed that a smaller fraction of raw material in geopolymerization allows for higher compressive strength [20].

Ilcan et al. [21] investigated the rheological properties of geopolymers based on construction and demolition waste as they applied to 3D printing technology. Geopolymer composites were made on the basis of 80% clay precursors, which are a mixture of hollow brick, red brick, and roof tiles; 10% concrete waste; and 10% glass. The mixtures were activated with various combinations of sodium hydroxide, solution, calcium hydroxide, and sodium silicate. The best compressive strength, tested after 28 days, was obtained for samples activated with a solution consisting of three activators (NaOH, Ca(OH)2, Na2SiO3)—36 MPa. The worst compressive strength was obtained for an alkaline activator based on a NaOH solution dissolved in tap water—11 MPa [21].

D’Angelo et al. [22] investigated the feasibility and potential of crushed brick waste (CBW) in the production of geopolymers to build precast components. Samples were obtained by curing a geopolymeric mixture at 60 °C for 3 days and aging for 28 days aging at room temperature. The results of the flexural and compression tests reached maximum values of 2.85 ± 0.73 MPa and 5.34 ± 0.66 MPa, respectively. The samples were deduced to have mechanical performance similar to that of gypsum produced from waste glass and ceramic waste. The conclusion was that CBW can be used successfully as a raw material in the construction of precast components [22].
Youssef et al. [23] examined the potential for reuse of waste brick (WB) by alkaline activation in a new geopolymer brick. Brick manufacturing was achieved by mixing WBs, ground granulate blast furnace slag (GGBFS), and sand with a solution of hydroxide and sodium silicate. The impact on properties based on variables was investigated, with the variables being the addition of GGBFS in different amounts, the molarity of sodium hydroxide (NaOH), and the silicate to sodium hydroxide ratio (Na$_2$SiO$_3$/NaOH). A maximum compressive strength of 89.91 MPa was obtained for a GGBFS/WB ratio of 80/20, an 8 M NaOH molarity, and a silicate/hydroxide ratio of 2/1. Comparably, for a GGBFS/WB ratio of 0/100 and analogous conditions, the compressive and flexural strength reached 38.96 MPa and 7.30 MPa, respectively [23].

In the literature, studies related to the development of geopolymers based on construction waste can be found, but to a much lesser extent than the commonly used aluminosilicates. This article uses research methods based on the analysis of the literature and the production of experimental geopolymer samples. Examples from world literature were analyzed in which the impact of replacing raw materials commonly used in the production of geopolymers with waste materials from building demolitions, replacing them completely in the mixtures produced, was analyzed. The authors of this article focused on the partial replacement of metakaolin and fly ash in geopolymers.

Despite previous research, still there is still a need for further research on the effective processing of CDW waste. It is worth mentioning that this type of waste requires recycling methods dedicated to particular geographical regions because of differences between used building materials, including raw material availability, building technology, and climate. The aim of this research is to use waste materials from building demolition to replace metakaolin and fly ash in geopolymer mixtures and to determine their mechanical and physical properties. To achieve the stated goal, ground clinker brick, concrete waste, metakaolin, fly ash, and a technical solution of sodium hydroxide with aqueous sodium silicate with a molarity of 10 M were used. The CDW from brick and concrete were mixed in GP to evaluate processing possibilities when the separation of waste is not possible. All the waste used came from south Poland. In the case of CDW, origin is quite important because of differences between the characteristics of the building materials used in different regions. The novelty aspect of the article is connected to the specific type of waste. This particular waste was not investigated before as a material for the geopolymerization process.

2. Materials and Methods
2.1. Materials

The research work used fly ash from the heat and power plant in Skawina (Poland), metakaolin from the Czech Republic (Keramost, Kadaň, Czech Republic), and clay brick from production waste (F.P.U.H. Cegielnia Kęty S.C, Kęty, Poland) and concrete debris. Clay bricks were from waste, and the piles have not been built in due to damage to the products. Concrete debris came from renovation work on building of the Faculty of Material Engineering and Physics (Cracow University of Technology, Cracow, Poland). Table 1 shows the percentage composition of the elements of individual components, determined by scanning electron microscopy with the energy dispersive spectroscopy (EDS) system for waste materials, clay brick, and concrete waste. EDS is a qualitative method, and based only on this research, it is not possible to obtain information about the quantitative composition of the material. However, this test confirms the similarity of this material to other investigated compositions and confirm the usefulness of this material to the geopolymerization process [17,19,22]. The chemical composition of fly ash (class F) contains less than 5% unburned carbon, less than 10% iron compounds, and a small amount of calcium. The amount of reactive silica is about 36% [24]. This type of fly ash contains many spherical particles [24]. Metakaolin contains many silicon oxides and aluminum oxides [25–27].
Table 1. Chemical composition of clay bricks and concrete waste.

| Element [Mass%] | O  | Al  | Si  | Fe  | K   | Na  | Mg  | Ca  |
|-----------------|----|-----|-----|-----|-----|-----|-----|-----|
| Clay brick      | balance | 7.11 | 30.6 | 6.2 | 1.7 | 0.8 | 0.7 | -   |
| Concrete waste  | balance | 1.9  | 11.8 | 2.0 | -   | 0.3 | 0.5 | 41.8|
| MK              | balance | 21.3 | 26.2 | 0.6 | 0.7 | -   | 0.1 | 0.3 |
| FA              | balance | 16.8 | 22.7 | 4.6 | 2.2 | 1.6 | 1.1 | 2.2 |

1 The other visible elements on EDS are Ti, S, P, Mn, and Cl (less than 1.0%). 2 The other elements, such as Ti, S, P, Ba, Mn, Cl, Sr, Cr, Zr, Zn, Cu, Pb, Ni, and Rb, are less than 1.6% [28].

Qualitative analysis of the elemental composition of the examined surfaces showed elements in clay brick and concrete waste (rubble) such as oxygen, silicon, aluminum, iron and—in the case of concrete waste—calcium. This is a typical composition for these types of materials. The elemental composition of clay brick is quite suitable for the geopolymerization process due to the reasonable amount of alumina and silica in the case of concrete debris. The amount of these elements is relatively low, but the composition is characterized by a large amount of calcium. There is a high chance that this raw material will work as a fine aggregate in the material rather than as a precursor for geopolymerization [24]. Because of that, it was decided to mix both wastes together.

The oxide composition for metakaolin and fly ash was determined. Table 2 shows the percentage composition of MK and FA. This composition shows a large amount of SiO$_2$ and Al$_2$O$_3$, which is an advantage for the geopolymerization process, and additionally a low amount of CaO, which is also advantageous from the point of view of creating the 2D structure of the geopolymer network [12]. Some amount of ferrous oxides—Fe$_x$O$_y$—was detected. This compound is mostly Fe$_2$O$_3$ [28,29].

Table 2. Oxide composition of the materials—MK and FA.

| Material | SiO$_2$ | Al$_2$O$_3$ | Fe$_x$O$_y$ | Na$_2$O | TiO$_2$ | K$_2$O | MgO | CaO |
|----------|---------|-------------|-------------|---------|---------|-------|-----|-----|
| MK       | 53.0    | 41.6        | 1.3         | 0.8     | 0.7     | 0.7   | 0.4 | 0.3 |
| FA       | 55.9    | 23.5        | 5.9         | 0.6     | 1.1     | 3.6   | 2.6 | 2.7 |

The particle distribution for clay brick and concrete waste used as raw materials is presented in Figure 1. The obtained values presented in the graph are the results of three measurements for each material carried out on the particle size analyzer. The analysis was carried out using a Particle Size Analyzer (AntonPaar GmbH, Graz, Austria).

In the case of clay bricks, 90% of the particles were less than 1240 µm and had a distribution width $D_{50}$ of about 480 µm. For concrete waste, 90% of the particles size were approximately 850 µm, with a distribution width $D_{50}$ of 425 µm. The mean sizes for these materials were approximately 560 µm and 460 µm, respectively. It is worth noticing that with a decrease in particle size, the mechanical properties of the geopolymer concrete increased. This is related to the greater surface area accessible for the reaction of synthesis [30]. For fly ash, 90% of particles were less than 30 µm, while their size ranges from 1.3 to 32.5 µm with a D$_{50}$ distribution width of 22.3 µm. On the other hand, 90% of the metakaolin particle size exceeded 30 µm; its particle size range was 0.5 µm to 39.5 µm, and the width of the distribution D$_{50}$ was 18 µm. This research was carried out in previous articles [30,31].
2.2. Samples Preparation

Two types of samples were produced as shown in Table 3. There are the same amounts of individual ingredients, the difference between them is the use of either metakaolin or fly ash.

| Sample | Metakaolin | Fly Ash | Clay Brick | Rubble |
|--------|------------|---------|------------|--------|
| G1     | 40         | -       | 30         | 30     |
| G2     | -          | 40      | 30         | 30     |

As an alkaline activator, a solution of technical sodium hydroxide flakes with aqueous sodium silicate (type R-145, density 1.45 g/cm³) in a ratio of 1:2.5 was used. Tap water was used to prepare the solution and mixed with sodium hydroxide flakes, and then sodium water glass (MR > 1.6–2.6) was added. The resulting solution was then left aside for 24 h until the component concentrations and temperature equilibrated. The appropriate amount of dry ingredients was placed in the bowl of the planetary mixer and mixed for 5 min. Then, a 10 M solution was added and mixed for another 10 min until the ingredients were combined. The obtained geopolymer mass was filled with a set of 50 mm × 50 mm × 200 mm prismatic forms and 50 mm × 50 mm × 50 mm cubic forms. The mold sets were then placed on a vibrating table to remove the air mass. They were then placed in a laboratory dryer for 24 h at a temperature of 75 °C. To prevent too rapid of a loss of the water, they were covered with a layer of polyethylene film. After this time, the samples were demolded and cured for 28 days at ambient temperature (ca. 23 °C). The next step was to determine the mechanical properties and microscopic observations.

2.3. Methods

The density of the produced geopolymers was determined by the geometric method on cubic samples. This method uses an electronic caliper (with an accuracy of 0.01 mm) and a RADWAG analytical balance (with an accuracy of 0.001/0.01 g). The density obtained is the average measurement result of six samples for each type.
The compressive strength of the geopolymer composites was determined in six 50 mm × 50 mm × 50 mm cubic samples using the MATEST 3000 kN test machine at a speed of 0.05 MPa/s according to EN 12390-3: Testing of hardened concrete [32].

The flexural strength was also performed on a MATEST 3000 kN testing machine with a speed of 0.05 MPa/s on samples of three 50 mm × 50 mm × 200 mm prismatic samples according to the EN 12390-5 standard [33]. The distance between the support points was 150 mm.

Pictures were taken using a JEOL JSM-IT200 scanning electron microscope with an energy dispersion X-ray spectroscopy system. Before microscopic images were taken, the samples were gold-plated for good conductivity using a DII-29030SCTR Smart Coater (JEOL, Tokyo, Japan).

3. Results

3.1. Density

Density values are shown in Table 4. The density was determined using the method described in Section 2.3 Methods. The measurements were performed on the samples before testing their mechanical properties.

Table 4. Density results of samples.

| Sample | Density [g/cm³] |
|--------|----------------|
| G1     | 1.81 (±0.13)   |
| G2     | 1.76 (±0.01)   |

Both samples have similar values for density. Higher-density geopolymers with densities of 1.81 g/cm³ have been obtained based on metakaolin and CDW. Geopolymer composites with fly ash as the base material had a slightly lower value—1.76 g/cm³. Density values for standard geopolymer created using a fine aggregate, usually sand, are typically between 1.30 and 1.80 g/cm³ [15,31]. The results obtained are similar to the higher value, typical for geopolymer concrete.

3.2. Mechanical Properties

3.2.1. Flexural Strength

The results obtained from the flexural strength tests for the samples are shown in Figure 2.

![Figure 2](image_url). Flexural strength (MPa) of the samples after 28 days of curing.
The samples for which metakaolin was used obtained better flexural strength—5.28 MPa. On the other hand, the samples containing fly ash as a main component achieved a compressive strength of 3 MPa, which is an acceptable value for many applications. The results obtained are comparable with other geopolymer composites based on fly ash and metakaolin [15,34].

3.2.2. Compressive Strength

The results of the compressive strength tests for both samples are shown in Figure 3.

![Figure 3. Compressive strength (MPa) of the samples after 28 days of curing.](image)

In the case of compressive strength, there is no statistical difference between the samples. The compressive strengths are: 20.1 MPa for samples based on metakaolin and 19.7 MPa for geopolymer composites based on fly ash. Both materials give similar values of compressive strength, which gives these materials the possibility to be applied in the building industry. Furthermore, these values are below the expected value for geopolymer concrete and are quite typical for geopolymer paste without fine aggregate [15,35]. However, similar values were also reported in the literature [34]. The lower values than expected are probably connected with the raw material. The amount of concrete debris used that can work as a fine aggregate is only 30%. In addition, the lack of proper activation can cause lower mechanical properties than expected [36,37]. Further research can be related to the increase in the amount of waste in the composition.

3.3. Microstructure Observations

The results of the SEM analysis are presented in Figures 4 and 5 at 500× and 1000× magnifications. The samples for the analysis were used after a compressive strength test, and because of this, some cracks are visible on the images.

In both cases, an amorphous structure is observed, characteristic of geopolymer materials, and the visible pores do not exceed 50 µm in size. Incoherent particles were not observed, which confirms the reaction between materials comprising the geopolymer mass. Analysis of sample G1 allowed sharp edges to be observed, while sample G2 was characterized by a less defined outline.

Additional information on the elemental composition was obtained from EDS. This analysis was carried out in the areas marked in Figure 4a for G1 and Figure 5a for G2. The results are presented in Table 5.
The elemental composition is typical for geopolymer material. The most common element in these material structures is oxygen. This is because the geopolymer material mainly consists of different types of oxides. In addition, significant amounts of silica and alumina are included in the material structure. They come from raw materials. These two elements are responsible for the formation of a proper geopolymer structure [11,12]. The small percentage of present silica could also be connected to the activator used. On the contrary, the presence of sodium is mainly related to the alkali activator. Only a
small amount of this element appears in the raw material. Potassium, magnesium, and calcium are also components of the waste used (Table 1). It should be noted that the EDS investigation has a qualitative character and cannot be treated as representative of these materials, so the differences between materials G1 and G2 cannot be treated as significant.

For the materials, elemental mapping analysis was also provided. The results are presented in Figures 6 and 7.

![Elemental mapping of G1](image1)

**Figure 6.** SEM images of sample G1 (a) elemental mapping; (b) whole spectrum.
Figure 6a shows the elemental mapping of the selected area for material G1 (composition based on metakaolin). Elements such as O, Na, Al, and Si have equal distribution in the whole area. The elements are easily visible because of the large amounts in the structure. Elements such as Mg, K, Ca, and Fe are not as well-represented in the microstructure. Some of them have a tendency to agglomerate at selected points.

In Figure 7a, the elemental mapping of the selected area for material G2 (composition based on fly ash) is presented. The distribution of the elements is similar to the composition G1. Large amounts of elements such as O, Na, Al, and Si are visible, as are less frequent appearances of K, Ca, and Fe. In the studied area, Mg is visible at only a few points in the material. The amount of Mg is lower than in the case of composition G1.

### Discussion

The results of prior research and the research carried out by the authors of this article prove the possibility of producing geopolymer materials using materials derived from the demolition waste of buildings and structures. A summary of the results of the exemplary literature can be found in Table 6.

**Table 6. Comparison of strength properties with literature results**

| Precursors | Activator | Compressive Strength after 7 Days [MPa] | Compressive Strength after 28 Days [MPa] | Flexural Strength [MPa] |
|------------|-----------|----------------------------------------|----------------------------------------|------------------------|
| G1         | 1 M (NaOH + water + sodium silicate) | 20.1 (±2.4)                           | 5.3 (±0.4)                             |                        |
|            | G2        | 19.7 (±1.0)                           | 3.0 (±0.4)                             |                        |
| Brick (CDW)| 8 M, 10 M (NaOH + water + sodium silicate) | 49.5                                   |                                        |                        |
| Tiles (CDW)| 8 M, 10 M (NaOH + water + sodium silicate) | 57.8                                   |                                        |                        |
| Concrete (CDW)| 8 M, 10 M (NaOH + water + sodium silicate) | 13.0                                   |                                        |                        |
| NGC (tile, red clay brick, hollow brick, glass, concrete waste, slag, fly ash, and natural aggregate) | 8 M (NaOH + water + calcium hydroxide + sodium silicate) | - 37.5 | 2.56 | 19 |
appearances of K, Ca, and Fe. In the studied area, Mg is visible at only a few points in the material. The amount of Mg is lower than in the case of composition G1.

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| Precursors | Activator | Compressive Strength after 7 Days [MPa] | Compressive Strength after 28 Days [MPa] | Flexural Strength [MPa] | Source |
|------------|-----------|----------------------------------------|----------------------------------------|------------------------|--------|
| G1         | 10 M (NaOH + water + sodium silicate) | -                                      | 20.1 (±2.4)                        | 5.3 (±0.4)              | current research |
| G2         | 10 M (NaOH + water + sodium silicate) | -                                      | 19.7 (±1.0)                        | 3.0 (±0.4)              | current research |
| Brick (CDW) | 8 M, 10 M (NaOH + water + sodium silicate) | -                                      | 49.5                                |                        |        |
| Tiles (CDW) | 8 M, 10 M (NaOH + water + sodium silicate) | -                                      | 57.8                                |                        | [20] |
| Concrete (CDW) | 8 M, 10 M (NaOH + water + sodium silicate) | -                                      | 13.0                                |                        |        |
| NGC (tile, red clay brick, hollow brick, glass, concrete waste, slag, fly ash, and natural aggregate) | 8 M (NaOH + water + calcium hydroxide + sodium silicate) | -                                      | 37.5                                | 2.56  |        |
| NGC—R (tile, red clay brick, hollow brick, glass, concrete waste, slag, fly ash, and recycled aggregate) | 8 M (NaOH + water + calcium hydroxide + sodium silicate) | -                                      | 36.6                                | 2.37  | [19] |
| 80% (hollow brick, red clay, roof tile), 10% glass, 10% concrete waste | NaOH + water | -                                      | 11.0                                |                        |        |
| 80% (hollow brick, red clay, roof tile), 10% glass, 10% concrete waste | NaOH + water + calcium hydroxide | -                                      | 17.9                                |                        | [21] |
| 80% (hollow brick, red clay, roof tile), 10% glass, 10% concrete waste | NaOH + water + calcium hydroxide + sodium silicate | -                                      | 36.0                                |                        |        |
| Clay brick wastes | 10 M (NaOH + water + sodium silicate) | -                                      | 5.34 (±0.66)                       | 2.85 (±0.73)  | [22] |
| 50% waste bricks, 50% sand | 8 M (NaOH + water + sodium silicate) | 38.96                                 | 7.30                                |                        | [23] |

The results show the possibility of using construction and demolition waste (CDW) as materials in the production of geopolymers. The mechanical properties are in the range of geopolymer materials received also from other authors (Table 6). In light of the research conducted, it is feasible to manufacture geopolymer composites using materials such as clay brick, concrete waste, roof tiles, or other elements of construction demolitions. The works provided demonstrate the potential of useful materials composed on industrial byproducts (fly ash) and CDW that provide environmental benefits [38,39]. This approach is important from the point of view of implementation of circular economy, which emphasizes the essence of using recycling materials, saving natural resources [40–42], and reducing the ecological impact of building materials [43,44]. Furthermore, the research highlights the possibility of using mixed CDW for the manufacturing of geopolymer concrete. The potential advantages of this approach are stressed in the literature because the separation of CDW is an easy process [45–47]. The literature indicates the prospect of manufacturing
this kind of material with reduced environmental impact, but there are still practical uses of it for the construction industry [48,49].

To reduce the impact of construction on the environment, it is necessary to search for new solutions and technologies that favor the development of a circular economy. The sustainable development of building materials is the main driving force behind research and application work in search of environmentally friendly products for use in construction. The production of geopolymers as materials to replace Portland cement leads to several environmental, economic, and social benefits: it reduces the amount of CO₂ released into the atmosphere [17]; allows the use of secondary raw materials, which reduces the use of natural resources; and lowers costs due to cheaper waste materials [50]. Due to the growing public awareness of activities that aim to reduce the impact of all types of materials on the environment, the area of geopolymers is still being developed [51]. An additional advantage of geopolymerization is the possibility of immobilizing various hazardous substances, thus securing waste landfills [52,53].

5. Conclusions

It is worth stressing that the topic undertaken in the article is important for the development of a circular economy, especially in the area of closing the loops in material recycling. The results obtained show that the elimination of CDW and the continual safe use of natural resources (raw materials), including the reduction of the impact on the environment and resource deficiency, is possible by the use of geopolymerization technology. In particular, the results of the research could find practical applications in the building industry. The use of waste materials such as CDW allows one to limit environmental impact, improve material circularity, and thereby bring benefits to society.

Two types of geopolymer composites were created on the basis of both a natural raw material—metakaolin—as well as a secondary raw material—fly ash—and waste materials—clay brick, concrete waste. Research on concrete cements included the determination of the strength properties in terms of partial replacement of the most commonly used materials (metakaolin, fly ash, volcanic tuff, slag) for the production of geopolymers. The results obtained show the possibility of manufacturing useful construction materials based on industrial byproducts (fly ash) and CDW. Based on the analysis of the particular research results, the following conclusions can be drawn.

- The densities of the materials were 1.76 g/cm³ and 1.81 g/cm³. These values are typical for solid geopolymer materials.
- The mechanical properties of the composites obtained are reasonable and allow them to be applied in the construction industry. The compressive strength and flexural strength were 20.1 MPa and 5.3 MPa, respectively, for samples with metakaolin additive. Geopolymers containing fly ash achieved 19.7 MPa of compressive strength and 3.0 MPa of flexural strength. The values obtained for flexural strength are typical for geopolymers. The compressive strength is below the preliminary expectation but comparable to the data obtained from the literature.
- SEM analysis provides useful insights into the mineralogy and microstructure of the produced geopolymers. This research shows the coherent and solid structure of the material obtained. The results of the EDS analysis are typical for geopolymer concrete.

To improve the properties of geopolymer concrete synthesized from byproducts and mixed CDW and next apply this type of composite to civil engineering, further research connected with other materials, such as fire resistance and other research, is necessary. In addition, optimization of material properties by applying desirable additives will be desired.

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References

1. EU Commission. Closing the Loop—An EU Action Plan for the Circular Economy. Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the Regions COM. 2015. Available online: https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:52015DC0614 (accessed on 23 March 2022).

2. EU Commission. A New Circular Economy Action Plan. Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the Regions COM. 2019. Available online: https://eur-lex.europa.eu/legal-content/EN/TXT/?qid=158933814386&uri=COM:2020:98:FIN (accessed on 23 March 2022).

3. Korniejenko, K.; Kozub, B.; Bąk, A.; Balamurugan, P.; Uthayakumar, M.; Furtos, G. Tackling the Circular Economy Challenges—Composites Recycling: Used Tyres, Wind Turbine Blades, and Solar Panels. J. Compos. Sci. 2021, 5, 243. [CrossRef]

4. Cerminara, G.; Cossu, R. 1.2—Waste Input to Landfills. In Solid Waste Landfilling, 1st ed.; Cossu, R., Stegmann, R., Eds.; Elsevier Science: Amsterdam, The Netherlands, 2018; pp. 15–39.

5. Chen, X.; Guo, Y.; Ding, S.; Zhang, H.; Xia, F.; Wang, J.; Zhou, M. Utilization of red mud in geopolymer-based pervious concrete with function of adsorption of heavy metal ions. J. Clean. Prod. 2019, 207, 789–800. [CrossRef]

6. Eurostat Yearbook: The Statistical Guide to Europe; Office for the Official Publications of the European Communities: Luxembourg, 2019. Available online: https://ec.europa.eu/eurostat/documents/3217494/10095393/KS-HA-19-001-EN-N.pdf/d434affa-99cd-4efb-a5e3-6d4a5f10bb07 (accessed on 20 May 2022).

7. Wang, R.; Li, S. Talking about the Production and Disposing of Construction Waste from the View of Sustainable Development. In Proceedings of the 2011 Asia-Pacific Power and Energy Engineering Conference, Wuhan, China, 25–28 March 2011; pp. 1–4.

8. Avilés-Palacios, C.; Rodríguez-Olalla, A. The Sustainability of Waste Management Models in Circular Economies. Sustainability 2021, 13, 7105. [CrossRef]

9. Dong, F.; Wang, Y.; Su, B.; Hua, Y.; Zhang, Y. The process of peak CO\textsubscript{2} emissions in developed economies: A perspective of industrialization and urbanization. Resour. Conserv. Recycl. 2019, 141, 61–75. [CrossRef]

10. Figiela, B.; Korniejenko, K. The possibility of using waste materials as raw materials for the production of geopolymers. Acta Innov. 2020, 36, 48–56. [CrossRef]

11. Davidovits, J. Properties of Geopolymer Cements. In Proceedings of the First International Conference on Alkaline Cements and Concretes, Kiev, Ukraine, 11–14 October 1994; pp. 131–149.

12. Davidovits, J. Geopolymer Chemistry and Applications, 4th ed.; Institut Geopolymere: Saint-Quentin, France, 2008.

13. Xu, H.; Van Deventer, J. The geopolymerisation of aluminosilicate minerals. Int. J. Miner. Process. 2000, 593, 247–266. [CrossRef]

14. Duxson, P.; Mallicoat, S.W.; Lukey, G.C.; Kriven, W.M.; van Deventer, J.S.J. The effect of alkali and Si/Al ratio on the development of mechanical properties of metakaolin-based geopolymers. Colloids Surf. A Physicochem. Eng. Asp. 2007, 292, 8–20. [CrossRef]

15. Bazan, P.; Kozub, B.; Lach, M.; Korniejenko, K. Evaluation of Hybrid Melamine and Steel Fiber Reinforced Geopolymers Composites. Materials 2020, 13, 5548. [CrossRef]

16. Lach, M. Geopolymer Foams—Will They Ever Become a Viable Alternative to Popular Insulation Materials?—A Critical Opinion. Materials 2021, 14, 3568. [CrossRef]

17. Şahin, O.; Içcan, H.; Ateşli, A.T.; Kul, A.; Yildirim, G.; Şahmaran, M. Construction and demolition waste-based geopolymers suited for use in 3-dimensional additive manufacturing. Cem. Concr. Compos. 2021, 121, 104088. [CrossRef]

18. Pacheco-Torgal, F.; Tam, V.; Labrincha, J.; Ding, Y.; Brito, J. (Eds.) Handbook of Recycled Concrete and Demolition Waste; Woodhead Publishing: Cambridge, UK, 2013.

19. Aldemir, A.; Akduman, S.; Koçer, O.; Aktepe, R.; Sahmaran, M.; Yildirim, G.; Almahmood, H.; Ashhour, A. Shear behaviour of reinforced construction and demolition waste-based geopolymer concrete beams. J. Build. Eng. 2022, 47, 103861. [CrossRef]

20. Komnitsas, K.; Zaharaki, D.; Velouchou, A.; Bartzas, G.; Galtakis, M. Effect of synthesis parameters on the quality of construction and demolition wastes (CDW) geopolymers. Adv. Powder Technol. 2015, 26, 368–376. [CrossRef]

21. Içcan, H.; Şahin, O.; Kul, A.; Yildirim, G.; Sahmaran, M. Rheological properties and compressive strength of construction and demolition waste-based geopolymers mortars for 3D-Printing. Constr. Build. Mater. 2022, 328, 127114. [CrossRef]

22. D’Angelo, G.; Fumo, M.; Merino, M.d.R.; Capasso, I.; Campanile, A.; Iucolano, F.; Caputo, D.; Liguori, B. Crushed Bricks: Demolition Waste as a Sustainable Raw Material for Geopolymers. Sustainability 2021, 13, 7572. [CrossRef]
23. Youssif, N.; Rabenantoandro, A.Z.; Dakhli, Z. Reuse of waste bricks: A new generation of geopolymer bricks. SN Appl. Sci. 2019, 1, 1252. [CrossRef]
24. Korniejenko, K.; Halyag, N.P.; Mucsi, G. Fly ash as a raw material for geopolymerisation—Chemical composition and physical properties. IOP Conf. Ser. Mater. Sci. Eng. 2019, 706, 012002. [CrossRef]
25. Ziefewski, C.; Marczyk, J.; Korniejenko, K.; Bednarz, S.; Sroczyk, P.; Lach, M.; Mikula, J.; Figiela, B.; Szeczyńska-Hebda, M.; Hebda. M. 3D Printing of Concrete-Geopolymer Hybrids. Materials 2022, 15, 2819. [CrossRef]
26. Walter, J.; Uthayakumar, M.; Balamurugan, P.; Mierzwiński, D. The Variable Frequency Conductivity of Geopolymers during the Long Agieng Period. Materials 2021, 14, 5648. [CrossRef]
27. Lach, M.; Korniejenko, K.; Hebdowska-Krupa, M.; Mikula, J. The Effect of Additives on the Properties of Metakaolin and Fly Ash Based Geopolymers. MATEC Web Conf. 2018, 163, 06005. [CrossRef]
28. Burduhos Nergis, D.D.; Vizureanu, P.; Sandu, A.V.; Burduhos Nergis, D.P.; Beijnariu, C. XRD and TG-DTA Study of New Phosphate-Based Geopolymers with Coal Ash or Metakaolin as Aluminosilicate Source and Mine Tailings Addition. Materials 2022, 15, 202. [CrossRef]
29. Sitarz, M.; Figiela, B.; Lach, M.; Korniejenko, K.; Mróz, K.; Castro-Gomes, J.; Hager, I. Mechanical Response of Geopolymer Foams to Heating—Managing Coal Gangue in Fire-Resistant Materials Technology. Energies 2022, 15, 3363. [CrossRef]
30. Korniejenko, K.; Kejzlar, P.; Louda, P. The Influence of the Material Structure on the Mechanical Properties of Geopolymer Composites Reinforced with Short Fibers Obtained with Additive Technologies. Int. J. Mol. Sci. 2022, 23, 2023. [CrossRef] [PubMed]
31. PN-EN 12390-3:2019-07; Testing Hardened Concrete—Part 3: Compressive Strength of Test Specimens. Polish Committee for Standardization: Warsaw, Poland, 2019.
32. PN-EN 12390-5:2019-08; Testing Hardened Concrete—Part 5: Flexural Strength of Test Specimens. Polish Committee for Standardization: Warsaw, Poland, 2019.
33. Marczyk, J.; Ziefewski, C.; Gadek, S.; Korniejenko, K.; Lach, M.; Góra, M.; Kurek, I.; Doğan-Sağlamtímur, N.; Hebda, M.; Szeczyńska-Hebda, M. Hybrid Materials Based on Fly Ash, Metakaolin, and Cement for 3D Printing. Materials 2021, 14, 6874. [CrossRef] [PubMed]
34. Kozub, B.; Bazan, P.; Gallitish, R.; Korniejenko, K.; Mierzwinski, D. Foamed Geopolymer Composites with the Addition of Glass Wool Waste. Materials 2021, 14, 4978. [CrossRef] [PubMed]
35. Figiela, B.; Simonová, H.; Korniejenko, K. State of the art, challenges, and emerging trends: Geopolymer composite reinforced by dispersed steel fibers. Rev. Adv. Mater. Sci. 2021, 61, 67. [CrossRef]
36. Korniejenko, K.; Lach, M.; Mikula, J. The Influence of Short Coir, Glass and Carbon Fibers on the Properties of Composites with Geopolymer Matrix. Materials 2021, 14, 4599. [CrossRef]
37. Wang, X.; Yang, W.; Liu, H.; Zhu, P.; Zong, N.; Feng, J. Strength and Microstructural Analysis of Geopolymer Prepared with Recycled Geopolymer Powder. J. Wuhan Univ. Technol. Mater. Sci. 2021, 36, 439–445. [CrossRef]
38. Huo, W.; Zhu, Z.; Chen, W.; Zhang, J.; Kang, Z.; Pu, S.; Wan, Y. Effect of synthesis parameters on the development of unconfined compressive strength of recycled concrete waste powder-based geopolymers. Constr. Build. Mater. 2021, 292, 123264. [CrossRef]
39. Das, S.K.; Shrivastava, S. Influence of molarity and alkali mixture ratio on ambient temperature cured waste cement concrete based geopolymer mortar. Constr. Build. Mater. 2021, 301, 124380. [CrossRef]
40. Moreno-Maroto, J.M.; Delgado-Plana, P.; Cabezas-Rodríguez, R.; Mejía de Gutiérrez, R.; Eliche-Quesada, D.; Pérez-Villarejo, L.; Galán-Arboledas, R.J.; Bueno, S. Alkaline activation of high-crystalline low-Al₂O₃ Construction and Demolition Wastes to obtain geopolymers. J. Clean. Prod. 2022, 158, 129770. [CrossRef]
41. Luhar, I.; Luhar, S.; Abdullah, M.M.A.B.; Nabialek, M.; Sandu, A.V.; Szmidtia, J.; Jurczyńska, A.; Razak, R.A.; Aziz, I.H.A.; Jamil, N.H.; et al. Assessment of the Suitability of Ceramic Waste in Geopolymer Composites: An Appraisal. Materials 2021, 14, 3279. [CrossRef] [PubMed]
42. Plawecka, K.; Figiela, B.; Grela, A.; Buczkoswska, K.E. Geopolymers based on plasma incineration waste as a material for circular economy. IOP Conf. Ser. Earth Environ. Sci. 2021, 942, 175470. [CrossRef]
43. Xu, Z.; Huang, Z.; Liu, C.; Deng, X.; Hui, D.; Deng, S. Research progress on mechanical properties of geopolymer recycled aggregate concrete. Rev. Adv. Mater. Sci. 2021, 60, 158–172. [CrossRef]
44. Alhawat, M.; Ashour, A.; Yildirim, G.; Aldemir, A.; Sahmaran, M. Properties of geopolymers sourced from construction and demolition waste: A review. J. Build. Eng. 2022, 50, 104104. [CrossRef]
45. Kvočka, D.; Lešek, A.; Knez, F.; Ducman, V.; Panizza, M.; Tsoutsis, C.; Bernardi, A. Life Cycle Assessment of Prefabricated Geopolymeric Façade Cladding Panels Made from Large Fractions of Recycled Construction and Demolition Waste. Materials 2020, 13, 3931. [CrossRef]
46. Ye, T.; Xiao, J.; Duan, Z.; Li, S. Geopolymers made of recycled brick and concrete powder—A critical review. Constr. Build. Mater. 2022, 330, 127232. [CrossRef]
47. Mahmoodi, O.; Siad, H.; Lachemi, M.; Dadsetan, S.; Sahmaran, M. Optimized application of ternary brick, ceramic and concrete wastes in sustainable high strength geopolymers. J. Clean. Prod. 2022, 338, 130650. [CrossRef]
48. Robayo-Salazar, R.; Valencia-Savedra, W.; Mejía de Gutiérrez, R. Recycling of concrete, ceramic, and masonry waste via alkaline activation: Obtaining and characterization of hybrid cements. J. Build. Eng. 2022, 46, 103698. [CrossRef]
49. Robayo-Salazar, R.A.; Valencia-Saavedra, W.; Ramírez-Benavides, S.; Mejía de Gutiérrez, R.; Orobio, A. Eco-House Prototype Constructed with Alkali-Activated Blocks: Material Production, Characterization, Design, Construction, and Environmental Impact. *Materials* 2021, 14, 1275. [CrossRef]

50. Davidovits, J. Environmentally Driven Geopolymer Cement Applications. In Proceedings of the Geopolymer 2002 Conference, Melbourne, Australia, 28–29 October 2002.

51. Bumanis, G.; Vitola, L.; Pundiene, I.; Sinka, M.; Bajare, D. Gypsum, Geopolymers, and Starch—Alternative Binders for Bio-Based Building Materials: A Review and Life-Cycle Assessment. *Sustainability* 2020, 12, 5666. [CrossRef]

52. Yao, Y.; Hu, M.; Di Maio, F.; Cucurachi, S. Life cycle assessment of 3D printing geo-polymer concrete an ex-ante study. *J. Ind. Ecol.* 2020, 24, 116–127. [CrossRef]

53. Kugler, F.; Aumüller, J.; Krcmar, W.; Teipel, U. Construction and Demolition Residuals as Raw Materials for the Production of Novel Geopolymer Building Materials. *Crystals* 2022, 12, 678. [CrossRef]