Experimental Investigation on the Development of Geopolymer Paving Blocks by Using Romanian Local Raw Materials

A Lăzărescu¹, H Szilagyi¹, C Baeră², A Hegyi¹ and V Meită³
¹NIRD URBAN-INCERC Cluj-Napoca Branch, Romania
²NIRD URBAN-INCERC Timișoara Branch, Romania
³NIRD URBAN-INCERC Bucharest, Romania

E-mail: adrian.lazarescu@incerc-cluj.ro

Abstract. For the world economic system cement and concrete are indispensable elements for the construction industry. Demand for concrete, hence for cement, is constantly growing, especially in highly developed countries, which means that alternative binders are urgently required to meet the needs of millions of people, without compromising the CO₂ levels of the atmosphere. Although environmental issues are currently not sufficiently convincing to create a sufficiently high demand for the production of geopolymer technology, this is expected to increase with the imposition of rules on carbon dioxide emissions and their effects. Preliminary alkali-activated geopolymer concrete mixtures were developed based on a rigorous trial-and-error process in order to establish the mechanical properties of the geopolymer paving blocks. The aim of this paper is to present results regarding the technology of development and optimisation for the production of geopolymer paving blocks, their mechanical properties and implementation possibilities in accordance with the intended scope of use. The study results indicate that alkali-activated geopolymer paving blocks have excellent mechanical properties, by reference to OPC paving blocks, making them suitable for practical applications.

1. Introduction

Worldwide, concrete is the second most used material and it is considered the most versatile, durable and reliable building material in civil engineering industry. Portland cement production is one of the most important carbon dioxide generators and global demand in the industry for its production as binder for concrete is rising constantly, generating environmental associated problems which are well known and start on being carefully monitored. Cement production requires large quantities of liquid fuel (between 60 and 130 kg / tonne) and the electricity amount required to produce on tonne is approximately 110 KWh [1]. In terms of environmental pollution, measurements have shown that for each tonne of cement that is produced, the industry releases into the atmosphere around 1.1 tonnes of CO₂, generated by the combustion and calcareous calcination methods [2]. World cement production indicates an obvious yearly increase, with more than 4.2 billion tonnes being produced and delivered to the industry only in 2014 [3].

A particular procedure in which the solidification the fly ash powder could be achieved is represented by its alkaline activation. When it is mixed with a certain type of alkaline activator, it creates a new binding material, with mechanical properties similar to those of Ordinary Portland Cement (OPC) concrete. Based on their properties, this type of new materials can be considered as an
alternative to OPC traditional concrete. Alkali-activated geopolymers are on-going developing materials, their research being strongly connected to the worldwide need of global CO\textsubscript{2} emissions reduction. Characterised by excellent mechanical properties and resilience in aggressive environments, these materials represent an opportunity for both the environment and the engineering fields of novelty as a reliable alternative to the traditional technology [4-6].

In order to produce alkali-activated geopolymer concrete, fly ash can be used as main raw, binding material. Therefore, careful consideration is needed when choosing this type of material to obtain the geopolymer binder. Depending on the type of coal that is burnt, the chemical properties of the fly ash can be very different and this has to be carefully considered when fly ash is used as raw material so that the geopolymerisation process can be fully generated [7]. The two main constituents of the geopolymer binders are the alumino-silicate elements, which represent the solid part, and the alkaline activator, which represents the liquid part [8].

Paving blocks have been used in the construction industry for many years, being considered appropriate for outdoor applications. These units turned out to be cheap to produce and present exceptionally high mechanical properties, making them useful for their intended purpose: commercial, industrial or residential areas, plazas, parking areas, bus stops, etc. In addition to being economical, interlocking concrete paving blocks are also broadly obtainable in water-permeable designs, which have additional ecological benefits [9].

Although the durability of alkali-activated geopolymer materials is not exactly extensively discussed and analysed, there are numerous studies regarding the performance of these materials in terms of sustainability [10-13]. Experimental results showed excellent resistance to aging, freeze-thaw, but also to carbonation [14].

The two main barriers to the introduction of new materials in the construction industry [15] are: 1) the need for standards for the testing methods, the evaluation and admissibility conditions, given that their development and introduction takes a long time and it is a gradual process; 2) the durability performance of such materials, taking into account the fact that they must meet certain requirements, they must withstand for a long, relevant period, the in situ (put in work), specific durability challenges, and for new materials this is not quantified yet.

The aim of this paper is to present results regarding the development technology and optimisation approach for the production of geopolymer paving blocks, by using Romanian local raw materials, their mechanical properties and implementation possibilities, in accordance with the intended scope of use. Based on the results obtained for alkali-activated geopolymer concrete [16], research directions on the material applicability were identified.

2. Materials and methods
This chapter summarizes the applied experimental methodology, involving the raw materials, mix design, moulding, curing and also the testing methods for the mechanical properties’ specific evaluation of the developed alkali-activated geopolymer paving blocks.

By using the full information on the production of the alkali-activated geopolymer concrete, as well as on the basis of the obtained results [6, 16], a “trial and error” experimental research was built, with the aim of producing building geopolymer materials (geopolymer paving blocks). The study of technology transfer to the industry, by enhancing the legal framework, which sets out the certification of products for the construction market, was also considered Geopolymer fly ash-based paving blocks were manufactured by the use of class F fly ash, aggregates and alkaline solution (sodium silicate + sodium hydroxide).

2.1. Fly ash
Low calcium-fly ash from a power plant in Romania was used as main raw material for the production of the alkali-activated geopolymer paving blocks. Its chemical composition was established through X-ray fluorescence methods (Table 1). As seen in Table 1, the CaO content of the fly ash was less than 10%
and SiO$_2$ + Al$_2$O$_3$ + Fe$_2$O$_3$ > 70%, SO$_3$ < 5% and L.O.I. < 6%. The chemical composition of the fly ash used in this study can be classified as Class F fly ash, according to ASTM requirements [17, 18].

**Table 1. Fly ash chemical composition.**

| Oxides | %   |
|--------|-----|
| SiO$_2$ | 46.94 |
| Al$_2$O$_3$ | 23.83 |
| Fe$_2$O$_3$ | 10.08 |
| CaO       | 10.72 |
| MgO       | 2.63 |
| SO$_3$    | 0.45 |
| Na$_2$O   | 0.62 |
| K$_2$O    | 1.65 |
| P$_2$O$_5$ | 0.25 |
| TiO$_2$   | 0.92 |
| Cr$_2$O$_3$ | 0.02 |
| Mn$_2$O$_3$ | 0.06 |
| ZnO       | 0.02 |
| SrO       | 0.03 |
| L.O.I.    | 2.11 |

2.2. **Alkaline activator**

A combination between sodium silicate solution (Na$_2$SiO$_3$ solution) and sodium hydroxide solution (NaOH solution) was chosen as alkaline activator for the production of the alkali-activated geopolymer paving blocks. The Na$_2$SiO$_3$ solution was commercially purchased and had the following chemical composition: SiO$_2$=30%, Na$_2$O=14% and H$_2$O=56%. Sodium hydroxide pearls were used to produce the NaOH solution, which was set to 10M, therefore 400g were dissolved in water, for one litre of solution. Based on the molecular weight of NaOH, in order to achieve the desired molar concentration of the solution 40g x 10 = 400g of NaOH flakes were dissolved in one liter of water.

2.3. **Aggregates**

Natural aggregates, granular class 0/4 mm (S) and 4/8 mm (CA) (sand and coarse aggregates) were used in this study to produce alkali-activated fly ash-based geopolymer concrete paving blocks.

2.4. **Mix design, moulding and curing treatment**

In order to study the mechanical properties of the alkali-activated geopolymer paving blocks, the following preliminary mixtures have been produced, considering the following parameters as constants: AA/FA ratio, Na$_2$SiO$_3$/NaOH solution ratio and S/CA ratio (Table 2).

The technology of producing alkali-activated geopolymer paving blocks (Figure 1) was based on the principles already set out for the production of alkali-activated geopolymer concrete [6, 15]. Mixing of the materials was performed at constant temperature of (20±2)°C and the following technological steps were applied:

1. Mixing of the sand and the coarse aggregates (0/4 mm and 4/8 mm) at low speed for 30 seconds;

2. The addition of the necessary amount of fly ash and then the dry raw materials (fly ash + aggregates) were mixing together, for 30 seconds;

3. The alkaline activator addition, for 90 seconds to the homogenous dry mixture;

4. After adding the total alkaline activator quantity, the mixing was continued at low speed for another 3 minutes;
(5) Casting into polypropylene moulds, with the corresponding 5 minutes vibration and starting of the heat treatment (70°C / 24 hours);

(6) After demoulding, the geopolymer paving blocks were stored in the climatic chamber at the temperature T (20±1)°C and relative humidity RH (60±5)%, and the tests for their mechanical properties were determined at the age of 7 days.

Table 2. Alkali-activated geopolymer paving blocks mix-design.

| Material / Parameter       | %   |
|----------------------------|-----|
| Fly ash                    | 1,0 |
| AA/FA                      | 0,9 |
| Na$_2$SiO$_3$              | 0,64|
| NaOH                       | 0,26|
| NaOH conc.                 | 10M |
| Na$_2$SiO$_3$ / NaOH       | 2,5 |
| S 0/4 mm                   | 0,93|
| CA 4/8 mm                  | 0,93|
| S : CA                     | 0,5 : 0,5|
| Binder / Aggregate         | 1 : 1,85|

Figure 1. Alkali-activated geopolymer paving blocks production.

2.5. Testing methods
Based on the results regarding the possibility of developing such construction elements [6, 15], the research initially focused on preliminary evaluation of the physical, mechanical and durability characteristics of the obtained products, for the study of their potential, different areas of applicability:

(1) Weathering resistance – total water absorption;
(2) Weathering resistance – freeze-thaw resistance with de-icing salt;
(3) Tensile splitting strength;
(4) Abrasion resistance;
(5) Slip/skid resistance;

All the tests were performed according to norm EN 1388:2004 / AC:2006: Concrete paving blocks. Requirements and test methods, which specifies the materials, characteristics, conditions and methods of testing the paving blocks, and it is applicable for the use of pedestrians, vehicles, bicycle lanes, parking lots, roads, highways, industrial areas, etc.. To be used in specific applications, they must comply with certain conditions at the time of their declaration as fit for use by the manufacturer.
3. Results and discussions
As mentioned in the literature, alkali-activated fly ash-based geopolymer materials properties are notably influenced by many important parameters. The physical and chemical properties of the main raw material (fly ash) used as precursor, the chemical properties of the solution used as alkaline activator and the mix-design ratios, are the most important parameters that affect the mechanical properties of the material [1, 4, 6, 10]. The 7 days compressive strength of the alkali-activated geopolymer paste (32.6 N/mm²) [16], produced using the same materials and the same mix-design ratios, was considered relevant as testing age for the evaluation of the above nominated parameters of the alkali-activated geopolymer paving blocks.

3.1. Weathering resistance
Weathering resistance for paving blocks is represented by the total water absorption test (according to Annex E from EN 1338:2004 / AC:2006) or the freeze-thaw resistance (according to Annex D from EN 1338:2004 / AC:2006). The recommendations related to the weathering resistance considered necessary to ensure the durability of the element, are determined by well-established criteria.

Total water absorption. Total water absorption of the alkali-activated geopolymer paving blocks has been determined by submerging the test samples in water, at constant temperature of (20±5)°C, until they reached constant mass. The minimum immersion period of the samples was 3 days. After reaching the saturated state constant mass, they were weighed (M₁). Subsequently, the paving blocks were placed in the oven and dried at temperature of (105±5)°C, to reach the constant mass (M₂). The Wa (water absorption) of each paving block was expressed according to the standard, as a percentage by mass, acc. to the equation (1) [19].

\[ Wa = \frac{M_1 - M_2}{M_2} \times 100 \]  
(1)

3.1.2. Freeze-thaw resistance with de-icing salt. The geopolymer paving blocks test specimens were preconditioned and then subjected to 28 freeze-thaw cycles, while their surface was covered with a 3% NaCl solution. After carrying out the 28 cycles, the exfoliated material was collected from the surface of the specimens (M) and the mass lost per unit area was calculated, acc to the equation (2) [18].

\[ L = \frac{M}{A} \, [kg/m^2] \]  
(2)

3.2. Böhme abrasion test
For the determination of this parameter 70 x 70 mm cubes were used, (Figure 2), cut from the geopolymer paving blocks and placed on the Böhme abrasive disc (Figure 3). Before the test and after every four cycles, the test specimens were weighted. Test specimens were tested for 16 cycles, each consisting of 22 full rotations of the abrasion track. After the end of the 16 cycles (Figure 4), abrasion was calculated [18].

3.3. Unpolished slip resistance (USVR)
Unpolished slip resistance of the geopolymer paving blocks has been determined by using a test equipment consisting of a friction pendulum (Figure 5), (Figure 6). At the pendulum swing (Figure 6), the force between the cursor and the test surface was measured by using a graduated scale. Prior the start of the test the geopolymer paving blocks were immersed in water at (20±2)°C for 30 minutes, according to the standard methodology requirements.

3.4. Tensile splitting strength
In order to determine the tensile splitting strength, the alkali-activated geopolymer paving blocks were placed in the testing equipment, in a special device for the tensile splitting strength test (Figure 7),
according to the specifications of the standard. The splitting load was applied progressively, until the paving blocks were broken (Figure 8).

According to EN 1338:2004 / AC:2006, the tensile splitting strength $T$ of the paving blocks should not be lower than 3.6 MPa, and the breaking load per length unit should not be lower than 250 N/mm [18]. Results obtained on geopolymer paving blocks show that the breaking load per length unit was 410 N/mm, and the tensile splitting strength was 3.6 MPa, which qualifies them as fulfilling the admissibility conditions specified by the assimilated standard.

Figure 2. Cubic specimen for the abrasion resistance evaluation.

Figure 3. Placing the cubic specimen in the testing abrasive machine.

Figure 4. Before and after, Bohme abrasion test specimen.

Figure 5. Slip resistance measuring equipment.

Figure 6. Unpolished Slip Resistance (USVR).
Figure 7. Loading conditions for the geopolymer paving blocks during the tensile splitting test.

Figure 8. Geopolymer paving block – failure mode after tensile splitting loading.

3.5. Mechanical properties of the alkali-activated geopolymer paving blocks

Alkali-activated geopolymer paving blocks have been subjected to tests for mechanical and durability performance evaluation by assimilating the methodology and admissibility conditions as well specified by standard EN 1338:2004 / AC: 2006. The obtained results on the nominated parameters confirmed their ability of the products to comply with this standard (Table 3), demonstrating the possibility of their use as paving elements in the construction materials industry. Moreover, by optimizing the mixes, improved performance can be achieved, and consequently the extension of the applicability interval within the specification of the EN 1338:2004 / AC: 2006.

Table 3. Geopolymer paving block characteristics.

| Mechanical property          | EN 1388 limit         | Results            |
|------------------------------|-----------------------|--------------------|
| Total water absorbtion       | ≤ 6 %                 | 5.6 %              |
| Freeze-thaw resistance       | ≤ 1.0 kg/m³           | 0.6 kg/m³          |
| Abrasion resistance          | ≤ 18000 mm³ / 5000 mm² | 16993 mm³ / 5000 mm² |
| Slip resistance              | -                     | 39 (moderate)      |
| Tensile splitting strength   | Min. 3.6 MPa          | 3.8 MPa            |
4. Conclusion and perspectives
Research regarding the production of alkali-activated geopolymer paving blocks using Romanian local raw materials could be definitely a possible answer to the need of greener concrete for sustainable development. Through the optimization of the alkali-activated geopolymer concrete mixtures, there was possible to demonstrate the viability of the material by producing geopolymer paving blocks and evaluating their mechanical and durability properties, thus opening up new opportunities for the future development of this type of material. One possible direction for the alkali-activated geopolymer paving blocks optimising process is related to psychological, mechanical and durability performance improvement by the addition of the natural fibres as disperse reinforcement [20].

Good mechanical properties, reached at very young ages (7 days) of the alkali-activated geopolymer binder, demonstrate the applicability of this material in the production of prefabricated construction products. Due to both, the raw material characteristics and the final properties of the material, the alkali-activated geopolymer paving blocks produced by using Romanian local raw materials showed all the test results within the admissibility limits imposed by EN 1388, hence demonstrating their possible use in residential driveways, light vehicles/public paths or other similar domain of use.

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