



Geopolymer Materials for Low-Pressure Injections in Coarse Grained Soil: Multiscale Approach to the Study of the Mechanical Behaviour and Environmental Impact

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Abstract The term soil improvement is commonly referred to the modification of soil structure in order to obtain a material with better physical and mechanical properties such as strength, stiffness or permeability. With this purpose, one of the most commonly used applications, particularly in coarse-grained soils, is the low pressure injection of cementitious mixtures. In recent years, there has been a growing demand for solutions with limited environmental impact and limited CO₂ emissions and, in this regard, the cement present in the injected grout is evidently the weak point of traditional solutions. In this work, the experimental study of geopolymer materials as a substitute of cement mixture for low-pressure injection for coarse-grained soils improvement is presented. The study started with a focus on the geopolymer fresh mixture properties (density, viscosity,

...) and the evolution over the time of the mechanical properties (compression and tensile strength and stiffness) comparing three different mix designs at three different monitoring temperatures. The same evaluations were repeated on sand samples injected with the different types of mixtures previously analyzed. For a selected mix design, a permeation test was carried out under controlled conditions to test the pumpability and effectiveness of geopolymer injection. Finally, to deepen the chemical interaction between the injected mixture and interstitial water, an injection test was carried out using a scaled model of a real injection system. The experimental study carried out was aimed both at the analysis of the characteristics of the geopolymer material and at its physical interaction with coarse-grained soil, passing through the measurement of the mechanical characteristics of the geopolymer material and of the solid sand skeleton mixed with geopolymers. Finally, the possible chemical interaction of the mixtures with groundwater was also evaluated in order to highlight any environmental issues. The results shown provide a preliminary but sufficiently broad picture of the behavior of geopolymer mixtures for low-pressure injection for coarse-grained soil improvement purposes both from physical–mechanical and chemical points of view.

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1 Introduction

Ground improvement techniques are widely adopted to modify soil mechanical and hydraulic properties as density, permeability, stiffness and strength (Puppala and Pedarla 2017).

In coarse-grained soils, due to their high permeability and relatively large pore size, it can be achieved by low-pressure injections of cementing-based grouts which, even partially filling the pores and maturing, create bonds between the granules/particles. Compared to the natural condition, the resulting material, therefore, is more rigid, more resistant and less permeable, since it is less porous. Most commonly adopted mixtures are in the form of suspension (one or more solid products dispersed evenly in water), solutions (homogeneous mixtures consisting of two or more components), emulsions (liquid, gaseous, or nanometric solid is homogeneously dispersed in another liquid phase) (Flora and Lirer, XXX) or resins (polyurethane, acrylic, etc.). These are very expensive materials, with a high environmental impact (to produce the raw materials relevant amounts of CO₂ are generated) and which, in some cases, provide very high final resistances of the treated soil, often even higher than necessary.

Several studies have been conducted with the aim of improving the in-situ properties of soils leading to the development of many methods, such as vibro-flotation, dynamic compaction, stone columns, jet grouting, compaction grouting. Many of these methods have limitations such as high cost and requirements of large equipment (Mutman and Kavak 2011). Low-pressure injections (permeation grouting), instead, requires simple injection apparatuses, lower upfront costs and easier on-site operations demonstrating to be a time/cost effective application (James Warner 2004). The technique applied for the treatment of coarse soil improves mechanical properties bonding the grains with each other by grout and reduces permeability reducing the volume of void originally containing water and/or air (S. et al. 2004; Lirer et al. 2006; Flora et al. 2006; Anagnostopoulos 2005). The grouting mixture is liquid, and it is usually injected into the soil by special pumps through pipes equipped with manchette valves (1–3 for meter) installed in the ground to be treated (Słowikowski and Kacprzak 2013). After the injection, the bonding of the grains starts and proceeds together with the hardening

process of the mixture. In such applications, the injection pressure plays a crucial role and its value must be selected to permit the injection of the grouting agent without modifying significantly the particles arrangement (Karol 1968; Donovan et al. 1984). For this purpose, it is possible to repeat the injections over time or increase the frequency of the injection to achieve a wide and as much as possible homogeneous/uniform treatment (Flora and Lirer, XXX). In practice, a pressure between 2 and 5 bar is used.

Examples of suspension mixtures are cement-based materials that guarantee significant increases in strength (Lirer et al. 2006). Although the advantage of obtaining more resistant and stiff materials, the use of these materials involves environmental issues. The cement industry is one of the major consumers of limited natural resources (water, sand, gravel) and contributes to about 10% of total greenhouse gas emissions, which corresponds to about 1.5 billion Gigajoules annually (Duarte et al. 2013; Feely et al. 2004; Hendriks et al. 2003; Humphreys and Mahasenan 2002; Spagnoli et al. 2021). For this reason, the research is directed towards the identification of alternative materials that are environmentally friendly without penalizing the mechanical performances.

In this particular perspective, geopolymers proved to be a good alternative to the use of cement-based mixture in geotechnical engineering applications, more specifically for low-pressure injection of coarse soil, leading generally acceptable results in terms of enhancement of mechanical properties and reduction of permeability (Nawaz et al. 2020). Moreover, geopolymers are known as eco-friendly materials because of the small emissions of CO₂ during their production cycle (Jiang et al. 2020) if compared to the greenhouse gas emissions due to the production of the cement (Duarte et al. 2013; Feely et al. 2004; Hendriks et al. 2003; Humphreys and Mahasenan 2002). Moreover, when industrial wastes (for example fly ash) are used as precursor material (Abdul Aleem and Arumairaj 2011; Farhan et al. 2020) it can also be defined as a sustainable material.

Going beyond general considerations: there is little information in the literature regarding the effectiveness of the use of these materials for low-pressure injection, there is a surprising variety of different materials and characteristics collected under the name of geopolymers, there are no systems of classification of mechanical properties, no specific studies have

been proposed to design specific injection interventions for geopolymer-based admixtures and doubts still exist on the environmental impact linked to the interaction of these admixtures with the environment (specifically with groundwater).

To contribute to reach a clearer picture of the performance of geopolymer materials, to analyse the interaction with coarse-grained soil and to provide data on releases into the water during the hardening process, in this paper the results of a laboratory investigation on the use of a commercial geopolymer material to be adopted in low-pressure injection in coarse-grained soil are presented. The followed approach permits the evaluation of mechanical improvement and environmental impacts of geopolymer injection in coarse-grained soil. In particular, the experimental programme was set up for: (i) evaluation of different mixture of geopolymer alone in terms of evolution of the mechanical properties over time; (ii) evaluation on its effect when mixed/injected with/into sand in controlled conditions; (iii) permeation test at small scale and controlled conditions to test injectability of the grout in coarse-grained soil and the effect in terms of variation of strength and stiffness of the soil and finally (iv) scaled model injection test to check the possible release of geopolymer in the interstitial water.

2 Materials and Methods

2.1 Geopolymers

Geopolymers are obtained from the dissolution and polycondensation of a silico-aluminate powder source activated by alkaline solution, generally constituted by potassium (the one used in the present research activity) or sodium silica, according to a step-by-step mechanism (Nawaz et al. 2020; Jiang et al. 2020; Abdul Aleem and Arumairaj 2011; Farhan et al. 2020; Medri 2009; Taki et al. 2020) that ends with the formation of a three-dimensional crystalline structure (Taki et al. 2020). The compressive strength of the geopolymers depends on reaction time. The percentage of water, added in the mixture to reduce viscosity and increase injectability, should be carefully selected because it strongly reduces/affects the mechanical properties (Jiang et al. 2020; Proia et al. 2017). On this point, Proia

et al. (2017) (Proia et al. 2017) performed research activities on nanosilica-based material showing that the characteristics of strength depend on reaction time reaching high values already after seven days and also noting that increasing the dilution with water reduces these characteristics of strength.

The temperature proved to be effective on the acceleration of solidification reaction which is highlighted by the increase of compressive strength of geopolymer specimens, especially in a range of temperature from 30 to 90 °C (Abdul Aleem and Arumairaj 2011). This is because high temperature enhances the complete dissolution of Si and Al species, speeds up the polycondensation process and, as a consequence, the achievement of the high level of strength of the geopolymer admixture (Nawaz et al. 2020; Farhan et al. 2020). On the other hand, high temperature, especially for a long time, could cause weakening of the micro-structure and reduction in compressive strength as a consequence of the water and alkaline solution evaporation from geopolymer admixture and an increase in porosity (Jiang et al. 2020; Farhan et al. 2020; Mo et al. 2014).

The geopolymer specimens were prepared from the dissolution of the precursor material, an aluminosilicate commercial powder (synthetic kaolin, $\rho_a = 0.55 \text{ g/cm}^3$), into a reactive alkaline potassium silicate solution ($\text{SiO}_2/\text{K}_2\text{O}$ weight ratio between 2.1 to 1.8, $\rho = 1.4 \text{ g/cm}^3$, $\text{pH} = 13$, Total Organic Carbon (TOC) equal to 450 mg/l). The materials are enriched with a micronized quartz ($\rho = 1.6\text{--}1.7 \text{ g/cm}^3$). In the following, as in engineering practice, it will be referred to the aluminosilicate powder as *precursor*, to the potassium silicate as *accelerator* and to the quartz powder as *filler*.

To give the injected mixture a higher viscosity, capable of being used even in the presence of large voids as well as to improve the performance of the hardened material, it is possible to add additives or fillers to precursor material and alkaline solution (Jiang et al. 2020). In this work, the quartz powder was used as a filler in all specimens.

The sand used for the preparation of sand/geopolymer specimens and for the simulation of injections comes from a quarry site near Colferro (Italy) with a grain size distribution reported in Fig. 1, a minimal void index $e_{\min} = 0.81$ and by maximum void index $e_{\max} = 1.11$.

Fig. 1 Grain size distribution of the sand from Colleferro used in the experimental activity

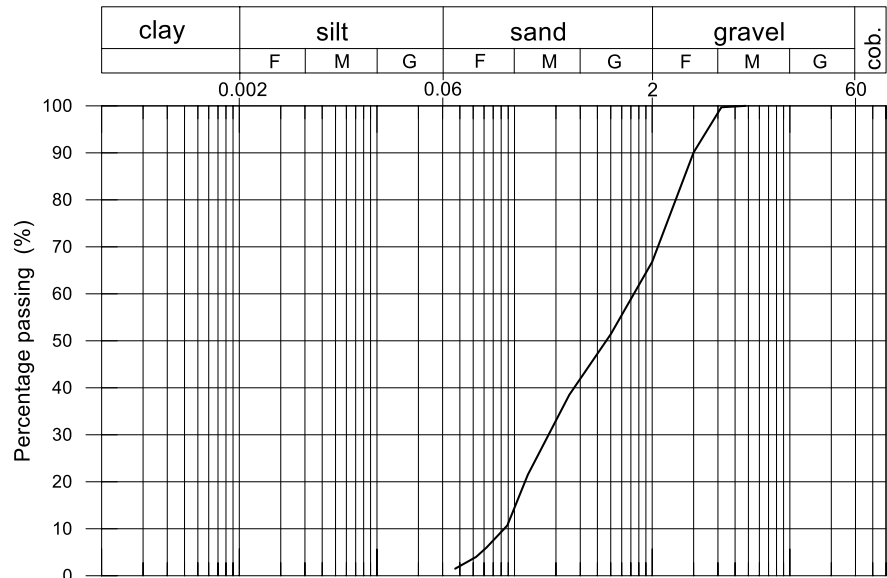


Table 1 Mix designs (% weight percentage)

MIX	M1 (%)	M2 (%)	M3 (%)
Aluminosilicate powder	31.9	28.4	24.3
Alkaline solution	28.7	25.6	21.8
Filler	31.9	28.4	24.3
Water	7.5	17.6	29.6

The mineralogical composition shows predominant percentage of Quartz (48%) and calcite 15%, albite 15%, microcline 13%, others 9%) (Guida et al. 2019).

2.2 Experimental Program

2.2.1 Preliminary Tests for Mix Design Selection and Geopolymer Mechanical Properties

As stated before, as the percentage of water increases the viscosity/injectability of the Mixture increases while the strength rapidly decreases. Thus, preliminary experiments were carried out to study the physical and mechanical behaviour of geopolymer alone. The objective of this study was the identification of the mixture to be adopted in the injections.

In detail, in the experimental program, the geopolymer mixture and filler were prepared according to the dosages reported in Table 1. The procedure

adopted for the production of geopolymer specimens involved firstly the mixing of precursor material and alkaline solution with a stirring mixer for 4 min at 800 rpm. Afterwards, water and filler were added a little at a time to the wet mix, and then mixed for two minutes. The fresh mixtures were then poured into cylindrical PVC moulds (diameter = 38 mm; high = 70 mm) to prepare cylindrical specimens that were cured for the first two days in the cylinders at a temperature of 25 °C. Starting from the third day, specimens were extruded placed into water to evaluate the possible release of species from the geopolymer for environmental investigations which will be discussed below. A further investigation was included by considering the effect of temperature (15 °C, 45 °C and 65 °C) on the mechanical properties. The viscosity of fresh mixtures was evaluated through the Marsh cone test.

Similar procedure was followed for mixture geopolymer-sand specimens preparation employing the three mixture reported in Table 1 with the addition of sand of about 38 g, 82 g and 135 g into the mixture M1, M2 and M3 respectively.

Samples of both geopolymer and geopolymer-sand mixtures, compressive strength tests were carried out after 18 h, 24 h, 7 days, 14 days, and 28 days from the preparation, and indirect tensile strength tests after 7 days, 14 days, and 28 days.

In Fig. 2, pictures of geopolymer and sand with geopolymer (prepared using M2 mix design) are presented.

2.2.2 Permeation Test at Small Scale and Controlled Conditions

To assess the injectability of the mixture, a necessary step before proceeding with the injection in a real system, a controlled injection was carried out inside a permeability cell. Taking into account the results obtained in terms of viscosity and strength on mixtures of geopolymer, the mix M2 was selected for the test, being the best compromise (relatively low viscosity and good strength).

The sand was inserted into the test cylinder and compacted in layers with a small weight. The mixture was then injected from 2 different injection points from bottom to top with a constant pressure of 2 bar.

After the injection, the coarse material resulted well permeated and homogeneously saturated with the geopolymer mix (Fig. 3). The sample was left to cure for 24 h inside the permeability cell, then extracted, covered with a wet cloth inside a plastic bag and finally placed to cure in a humid environment at the same for environmental investigation. After 28 days, selected as maximum time for the completion of polycondensation reactions and for the monitoring of species that slip to geopolymer formation, the specimen was cut to obtain two slices of about 1.5 cm thickness and a cylinder. The cylindrical



Fig. 3 Cylindrical sample

specimen was subsequently subjected to a uniaxial compression test in order to evaluate the compressive strength while on the two disks indirect tensile tests were carried out.

2.2.3 Injection Test on Scaled Model

The optimal mix design individuate through preliminary tests results was employed for the simulation

Fig. 2 Specimens: **a** geopolymer alone; **b** mixture of sand and geopolymer



(a)



(b)

of a real injection using a scaled model to deeply analyse the interaction between geopolymer and water during the condensation in one configuration similar to the in-situ situation. The mixture injected was prepared according to the procedure previously reported. In the first part of this study the design of the injection system to simulate the low-pressure injection process was addressed. In Fig. 4a–d is reported the schematic representation of the system conceived and built-in laboratory scale dimension. The injection system here proposed was realized in the laboratory and allowed two different injections; it is composed of an injection probe (Fig. 4b), a metal tube positioned at different heights (Fig. 4c), a probe with four holes for the passage of the mixture and two rubber rings (Fig. 4d).

The injection tube was fixed in a cylindrical container, filled with sand ($d > 1$ mm). The injection, realized at 2 bar of pressure, was performed at different levels by modifying the position of the probe along the perforated tube.

2.3 Methods

2.3.1 Mechanical Characterization

To evaluate the mechanical properties of the specimens of geopolymer and geopolymer/sand admixture, uniaxial compressive strength and indirect tensile strength tests were carried out.

The UCS tests were carried out according to AS 1012.9-1999 (“AS 1012.9, Methods of Testing Concrete; Method 9: Determination of the Compressive Strength of Concrete Specimens” 1999) with a displacement rate of 0.5 mm/min. Stress and deformation data were recorded through an electronic system. Before testing, the specimens were smoothed to ensure a uniform loading surface.

Indirect tensile strength tests were carried out according to AS 1012.11-2000 (“AS 1012.10, Methods of Testing Concrete—Determination of Indirect Tensile Strength of Concrete Cylinders Brasil or Splitting Test.” 2000). The tests were conducted at the same displacement rate as the UCS tests.

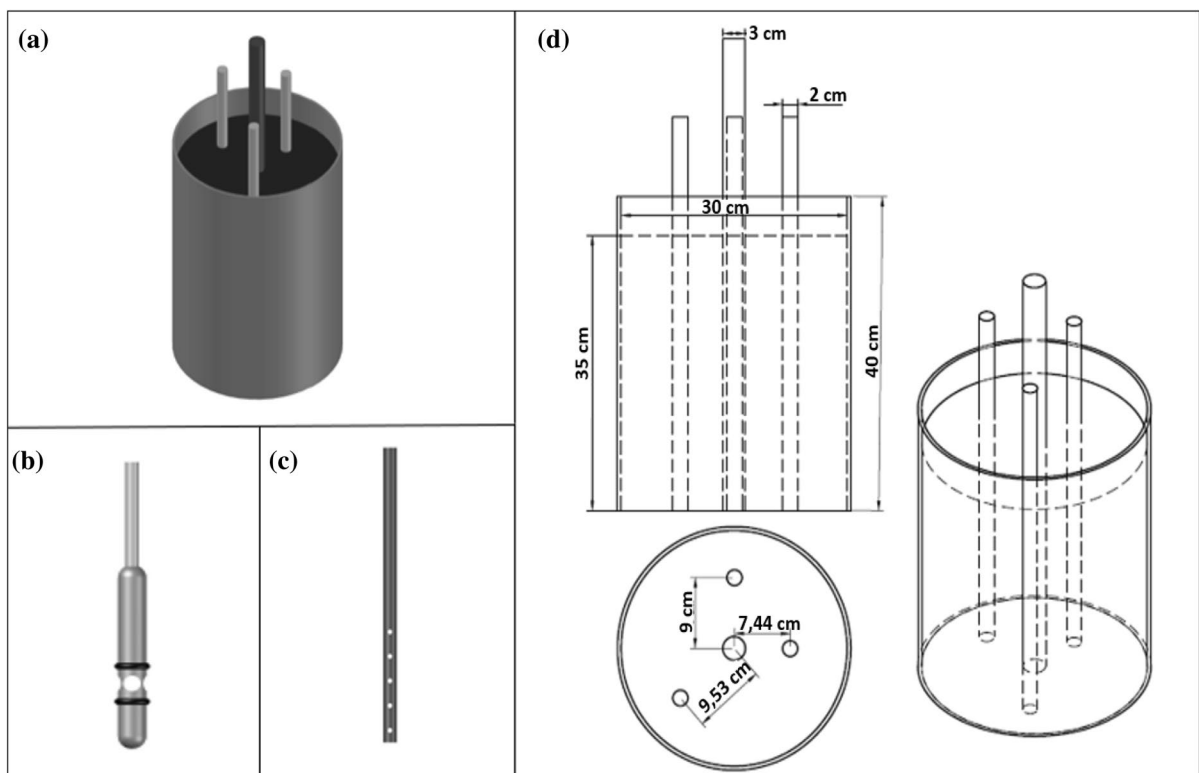


Fig. 4 Model: **a** cylindrical case, **b** injection system, **c** drilled tube and **d** details and sizes of the system

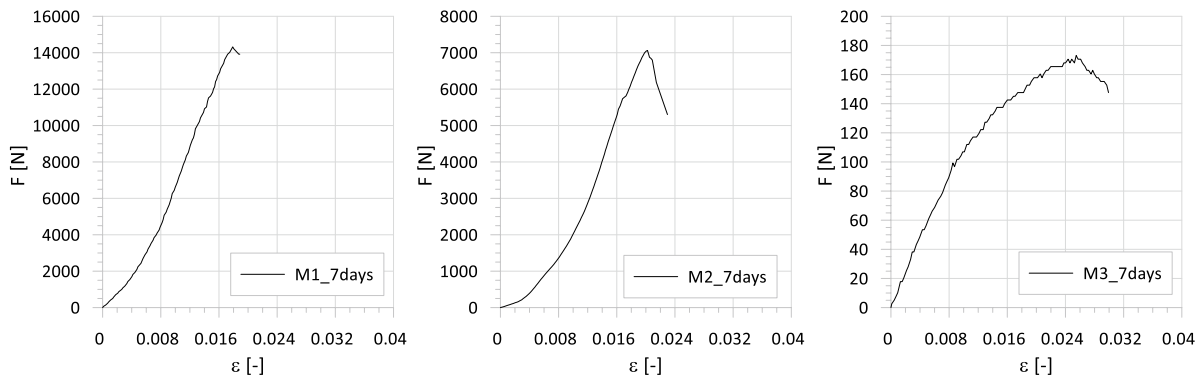


Fig. 5 Unconfined compression tests on M1, M2 and M3 samples at 7 days

2.3.2 Chemical Characterization

To evaluate the environment impact of geopolymer-based mixtures materials, a detailed analysis of water solutions used for the storage of the specimens during the preliminary test investigations and the interstitial water taken from the sample prepared for the mesoscale permeation test is presented. Total Organic Carbon (TOC, TOC-Analyzer Shimadzu) and pH (pH-metro Crison GLP 22) were measured. SiO₂ concentration was detected by spectrophotometer analysis using a PG Instruments T80 + UV/Vis spectrophotometer (using a quartz cell of 1 cm path length) according to Standard Method Silica (4500-Si)/Molybdosilicate Method.

3 Results and Discussion

3.1 Preliminary Test on Geopolymer Alone

Before the mechanical properties investigation, the selection of the mix design range adopted in this work was supported by the measure of the viscosity of fresh mixtures (Table 1) through the Marsh cone test. Values of 2160 s, 102 s and 34 s were measured for Mix1, Mix2 and Mix3, respectively. The mixture M3 prepared with higher percentage of water (29.6%) resulted extremely liquid; while using lower percentage of water (7.5%), the Mix1 specimens resulted hardly workable and very difficult to inject because its very high viscosity.

In Fig. 6 the results of UCS tests and indirect tensile strength test performed on the same samples

described in par. 2.2.1, are reported. All samples were tested after different time from 18 h to 28 days. As regarding the compressive strength, for all mixtures the same trend was observed: an increase of UCS values was measured up to 14 days, after that the UCS trends revealed a plateau. This is particularly evident for geopolymer specimens M1 and M2. This evidence highlights that to complete the dissolution and poly-condensation reactions are necessary at least 14 d.

Furthermore, from the trend of the curves it is possible to see how the mechanical behavior of the specimens changes as the monitoring time increases: the specimens of at 18 h and 24 h show a ductile behavior, while at 7 days and 28 days the behavior became fragile with a markedly strength reduction after a peak. This behavior, as expected, is typical of rocks and more in details the recorded value in term of strength and stiffness are comparable to weak rocks.

It is worth noting that the compressive strength depends significantly on the amount of water in the mixture and this justifies the lowest compressive strength resistance (Fig. 5a) measured in case of M3 attributable to the highest water content (Table 1).

The tensile strength showed a trend similar to those observed for UCS. In fact, it is possible to notice an increase of strength along time and a plateau after 14 days (Fig. 6b). The ratio between tensile strength and compressive strength at 28 days resulted equal to 0.1, 0.1 and 0.06 for M1, M2 and M3, respectively. Once again, such values are similar to those of rocks.

To complete the investigation, the chemical composition of water solutions in contact with specimens was detected. The pH values of the aqueous solutions were in the range of 11.8 and 12.4. This because of

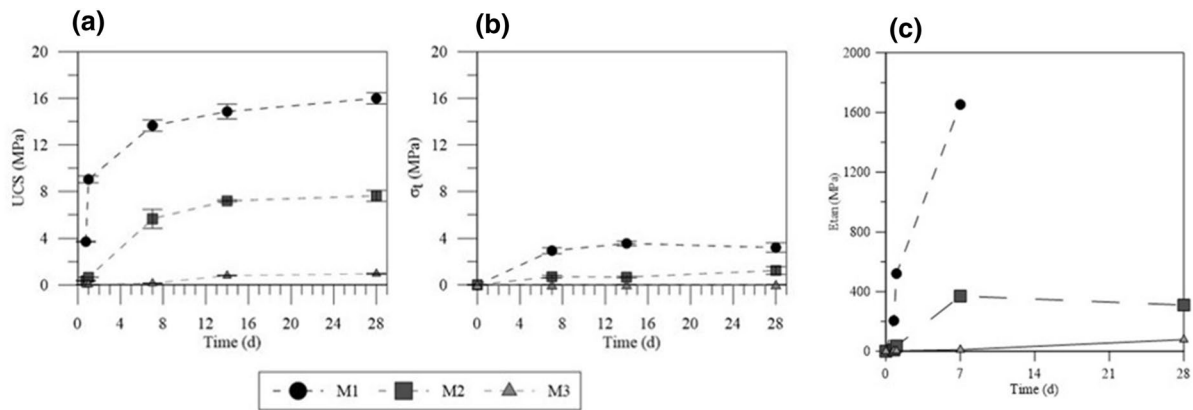


Fig. 6 Mechanical tests result for mixture with geopolymer alone: **a** and UCS test **b** indirect tensile strength test **c** evolution over the time of the stiffness of the samples

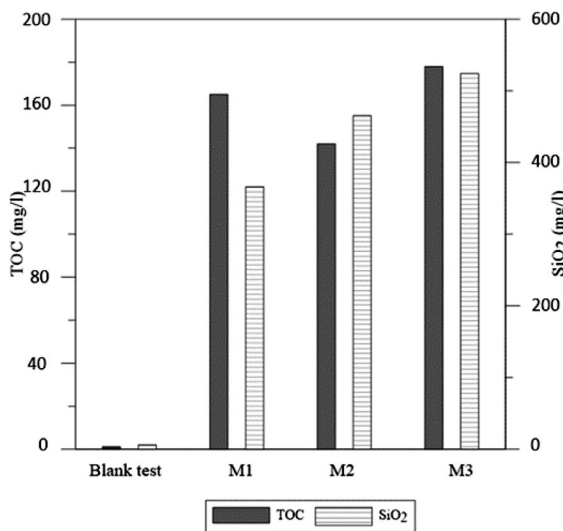


Fig. 7 TOC e SiO₂ concentrations on water in contact with specimens after 28 days

the presence of the liquid alkaline solution ($pH=13$) in all mix designs. As regarding the chemical composition, the release in water solution is mainly attributable to TOC (alkaline activator) and SiO₂ species as reported in Fig. 7. For this reason, the concentration of organic compounds and silica species in water solutions are reported (Fig. 6). All water samples revealed a significant concentration of both species with respect to the blank (water composition before the contact with solid samples). As regarding the TOC values, a low value for M2 with respect to M1

was observed and this because of the low precursor content in M2 respect to the mix design of M1 (Table 1). While an opposite trend was found with a low concentration of alkaline precursor in M3. The release observed in the case of M3 was amplified by the delay in polycondensation process.

As regarding the release of silica, their dissolution in water was mainly due to the different times required for the formation of the crystalline structure and as a consequence, the delay observed in the case of M3 mix design is reflected in a major dissolution of this species in the water phase. This suggests that, until the polycondensation reaction is completed such substances may remain in the liquid phase (water) as a consequence of dissolution step.

This consideration is also in line with the evidence of UCS (Fig. 6a), the remarkable differences in mechanical resistance were a consequence of two contributes the increase of water content in the mix design from M1 to M3 and the silica dissolution enhanced by the delay on harden process. As regarding the solid matrix, X-ray Powder Diffraction analysis (data not reported) revealed that the geopolymer was mainly based on quartz.

It is known that the operative temperature has an effect on the geopolymer hardening process (Mo et al. 2014) and with the aim to define the optimal temperature condition and the trend of UCS values, different tests were done with specimens characterized by M2 as mix design.

How is possible to observe in Fig. 8, the change in temperature leads to an increase in

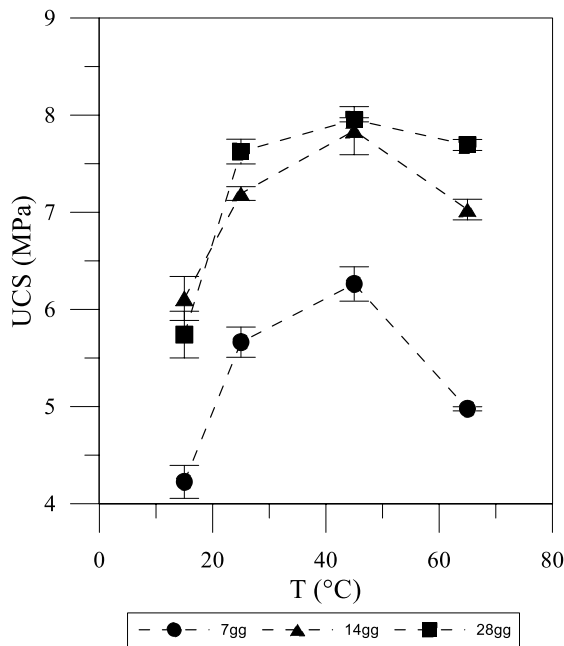


Fig. 8 UCS test results at different temperatures for the mix design M2

the mechanical behavior up to 45 °C. A further increase in temperature proved to be negative for the mechanical resistance development. According to what reported in the literature, high temperature values contribute to the evaporation of water and alkaline solution, leading to incomplete reactions and the formation of pores inside the specimens (Jiang et al. 2020)(Farhan et al. 2020).

The greatest development of resistance was observed in the period between 7 and 14 days, while up to 28 days there was a very small enhancement on mechanical resistance and the values become almost overlapping, suggesting that the polycondensation ends in the first 14 days. This consideration is very important when it is necessary to select a specific mix design to prepare materials with certain requirements in terms of mechanical resistance. 14-day was sufficient to assess the complete development of resistance that, in the case of 25 °C as operative temperature, corresponds to 91% of the maximum UCS measured in case of 45 °C as temperature. As regarding the TOC, the temperature did not influence significantly the release of organic

compounds in the water solution that was measured on average equal to 151.47 ± 8.49 mg/L.

From a geotechnical point of view, the results observed for the test at 15 °C can be considered a simulation of what happens during the real injection in soil. In fact, this process is operated at a temperature lower than the room temperature and, in such conditions, the polymerization reaction is not triggered and all the bonds typical to the geopolymer structure will not be created in a short time. Also in this case, 14 days were necessary to observe and measure the effect of soil improvement. The selection of an appropriate temperature for laboratory-scale investigations is closely dependent on in-situ expected conditions.

3.2 Tests on Sand Injected with Geopolymers

A similar investigation was conducted on samples prepared with sand ($d < 1$ mm) with the addition of geopolymer admixture. UCS tests were carried out after 18 h, 24 h, 7 days, 14 days, and 28 days. The results showed in Fig. 9a have the same trend observed for the results of specimens of geopolymers alone. In both cases, the measured strength obtained employing M1 and M2 are similar with those of soft rocks; while the strength obtained with M3 are quite low.

Moreover, referring to the use of M2 mixture, considered the best compromise between material injectability and mechanical properties, a comparison between the compressive strength results (Fig. 9), shows, irrespective of curing time, that the strength of geopolymer alone (Micro0) is similar with that of sand mixed with geopolymer (Micro1).

As regarding the TOC release of specimens, in Fig. 10 a comparison between the TOC values of casting water solution of geopolymer and geopolymer and sand samples is proposed. The co-presence of geopolymer and sand not determined a significant modification of TOC as expected the organic source was identified by the alkaline activator used for the geopolymer formation. Similar results to geopolymer alone analysis were observed in the case of SiO_2 release for sand-geopolymer mixture. This confirms that, in the case of a large-scale application, environmental monitoring has to be defined in order to evaluate the impact of TOC and SiO_2 species into

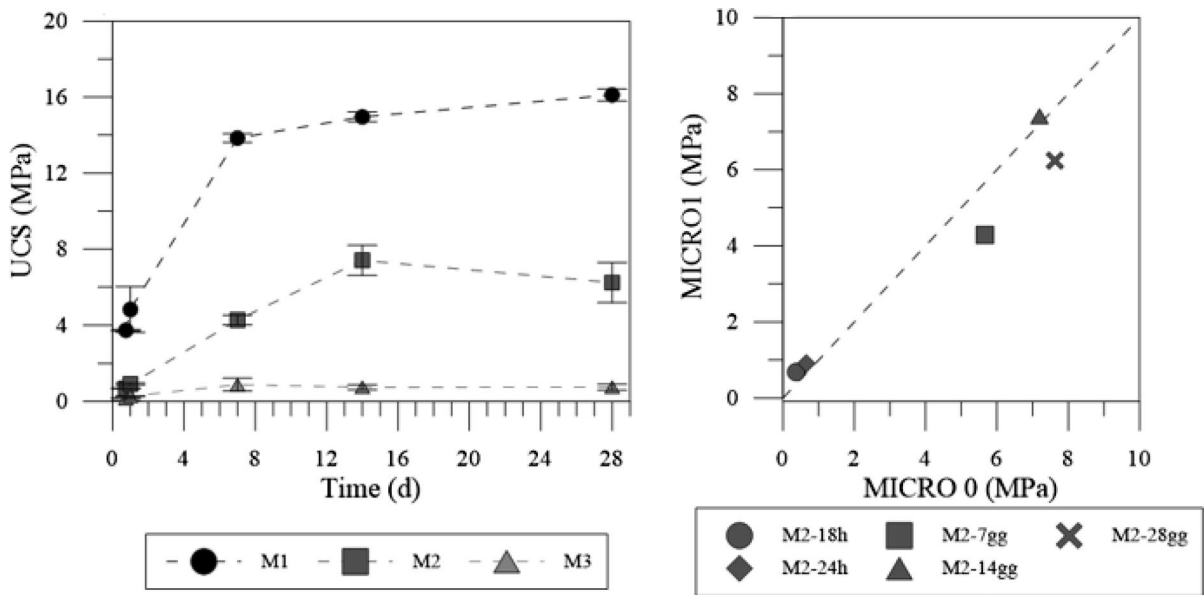


Fig. 9 Mechanical test result on small scale samples of geopolymer and sand injected: **a** UCS test results on sand-geopolymer mixture; **b** comparison between compressive strength of

specimens of geopolymer alone (Micro0) and sand+geopolymer mixture (Micro1)

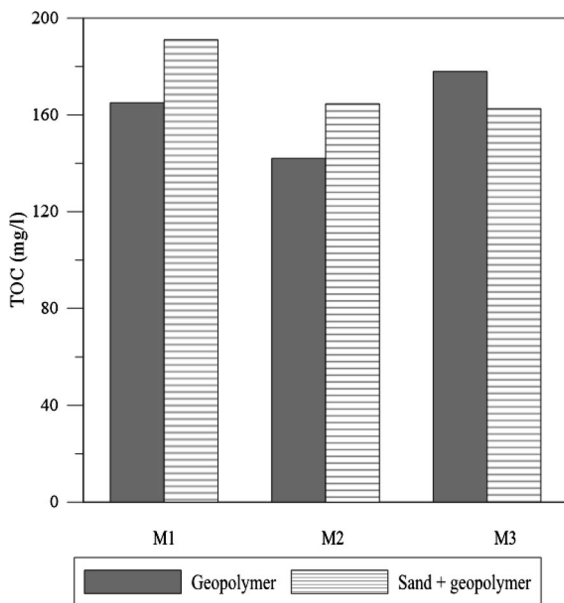


Fig. 10 TOC analysis of water solutions after 28 days of specimens curing for 28 days

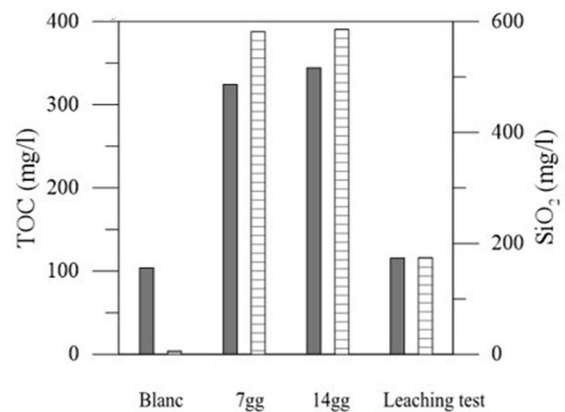


Fig. 11 Mesoscale results: TOC and SiO₂ releases in water solution

the consolidated soil and the possible contamination effects of the interstitial aqueous phase.

3.3 Tests on Scaled Model

The optimal M2 mixture was adopted for the injection in the scaled model previously described (§. 2.2.3, Fig. 4). The mixture was injected starting from the lowest hole in the tube with an injection pressure of 2 bar.

Regarding the monitoring of environmental parameter in the soil and water phase, the analysis

of the interstitial water phase (Fig. 11), in a time of 7 days and 14 days after the injection, revealed that organic compound and SiO_2 were released in few days in the water phase as a consequence to their high solubility. Of course, the high numerical values are a consequence of the accumulation of such species in a very limited volume of water. After 28 days a leaching test of the soil was done and the release of compound accumulated in the solid matrix was compared with the results obtained with the leaching test operation on the soil before the injection. In this case, the appreciable TOC value can be considered related to possible organic contaminants or substances present in the soil and the addition of geopolymer not revealed an alteration of this parameter. On the contrary, the nature of the geopolymer admixture (silico-aluminate based) involved an alteration in terms of silica release.

4 Conclusions

The results of an experimental investigation on mechanical behaviour of geopolymer materials used for low-pressure injection for soil improvement were presented. The main aim of this research was the identification of a mix design capable of ensuring good injectability (proper viscosity) and good mechanical properties (strength and stiffness).

The experimental program included: (1) tests required to define the mix design and the mechanical properties of the geopolymer grout to be injected and the properties of sand samples mixed with this grout; (2) a permeation test under controlled conditions performed to verify the injectability of the grout; (3) injection test in a scaled model performed to analyse the release of chemical compounds into the interstitial water.

The most relevant results obtained in this study can be summarized as follow:

- The selected polymeric mixture (precursor + accelerator + filler) in all three different studied mix designs is able, over the time, to develop a crystalline structure;
- The physical and mechanical characteristics of the prepared mix designs strongly depend on the percentage of water used. Stiffness and strength significantly decrease as the percentage of water

increases. The viscosity of the freshly prepared mixture significantly decrease as the water content increases;

- The polycondensation takes place quite quickly and, for these mixes, can be considered completed in 14 days;
- Curing temperature plays an important role for all selected mix designs; in particular, stiffness and strength increase as temperature increases from 15 °C to 25 °C and then to 45 °C; on the opposite, both decrease sharply in the tests performed at 65 °C;
- The mixture M2 selected as most suitable for the purposes of the work (treatment by low pressure permeation of coarse-grained soils) showed a good compromise between injectability and mechanical properties (more than adequate for the purpose);
- The good injectability of this mixture has been experimentally demonstrated achieving the saturation by permeation of a sand sample under low pressures;
- It seems that the geopolymer injected in the sand continues to react over the time, producing a three-dimensional structure that adheres to the granules and connects them with strong cementing bonds;
- In the case of direct injection in soil (scaled model), the analysis of the interstitial water phase, at 7 days and 14 days after the injection operation, revealed the presence of organic compound (TOC) and SiO_2 as a consequence to their high solubility; after a maximum time of 28 days a leaching test of the soil was done and the release of compound accumulated in the solid matrix was compared with the results obtained with those of the soil before the grouting operation. In this case, the appreciable TOC value can be associated to organic contaminants or substances present in the soil, and the addition of geopolymer not revealed an alteration of this parameter. On the contrary, the nature of the geopolymer admixture (silico-aluminate based) involved an alteration in terms of silica that not contributed to geopolymer formation;
- In addition, the concentration of chemical compounds as TOC and SiO_2 in water were not strongly influenced by the mix design adopted but due to the in-situ geopolymerization. The concentration of such species must be monitored to pre-

vent any adverse effect in term of soil/groundwater contamination.

Finally, the geopolymer material studied (precursor+accelerator+filler) in the selected mix design has good characteristics of strength/stiffness and injectability and is therefore a candidate to replace the cement-based grouts and other chemical-based alternatives for the improvement of the mechanical characteristics and to reduce the permeability of coarse-grained soils by permeation at low pressure.

As a further development of the presented study, primarily chemical and mineralogical test will be performed to detect the mineralogical structures and minerals resulting from the formation of the polymeric materials. Moreover, a real scale pilot injection test is recommended to verify injectability and the environmental releases in real scale and conditions.

Author contributions All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by MB and CC. The draft of the manuscript was written by IB and DS, reviewed by ADG, LDP, SM and QN. All authors read and approved the final manuscript.

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