

Powder bed 3D printing with quarry waste

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Abstract. With 3D printing, material consumption can be reduced: It allows to place material only where needed, therefore structurally optimized building parts or formworks can be printed. Currently this technique uses an epoxy based resin to glue layer by layer an inert sand bed. However, this material choice holds a large improvement potential from an environmental and health perspective. It was shown previously, that the organic glue, that releases unhealthy volatile organic compounds, can be replaced by a mineralic binder, namely a geopolymer. With geopolymers and alkali activated materials, the embodied energy can be reduced, especially when built from waste materials. In this study, we focus on the replacement of the sand, which is becoming a scarce resource. The waste from a local gneiss quarry in Ticino (Switzerland) could be a good alternative. The powder bed of the 3D printing is made of aggregates of crushed quarry waste mixed with an aluminosilicate powder. The printing liquid is an alkaline solution that activates the aluminosilicate and reacts to a geopolymer. Droplet penetration experiments on different powder mixes were performed to adjust the binder composition. With a custom built powder bed 3D printer, samples with varying compositions and porosities were printed and tested on compression. It could be shown that samples printed with quarry waste perform as well as samples printed with silica sand in terms of compression strength and accuracy. This new material system is promising: with 3D printing of geopolymers and quarry waste, we can combine the environmental benefits of a new building technique with a low carbon intense material. The application of this technique may help the sustainable development of the local quarry sector by consuming the volumes of waste that causes storing and ecological issues and keep small quarries running.

1. Introduction

The construction industry consumes more than 40% of the resources available worldwide [1], and causes huge CO₂ emissions. Construction resources such as gravel and sand are becoming scarce in some places [2, 3]. To face these problems we need to develop construction techniques which combine a reduced resource consumption with lower greenhouse gas emissions. An upcoming technique in construction is 3D printing. Indeed, it can help to reduce the material consumption: with 3D printing material is placed only where needed. Hence, structurally optimized building parts or formworks can be printed [4, 5].

There are many different 3D printing techniques such as fused deposition modelling (FDM), selective laser sintering (SLS), stereolithography (SLA) or binder jet 3D printing. Among these, FDM is the most popular in construction [6]. A paste material such as concrete can easily be extruded, but the

main drawback of FDM is the limited overhang printing. This limitation can be overcome by powder bed binder jetting [7].

The principle of binder jet 3D printing is explained in Figure 1: a liquid binding agent is deposited to join powder particles previously placed on a movable platform. Another layer of powder is then spread on top, and more binder is added. Over time, layers of material are bonded to form an object.

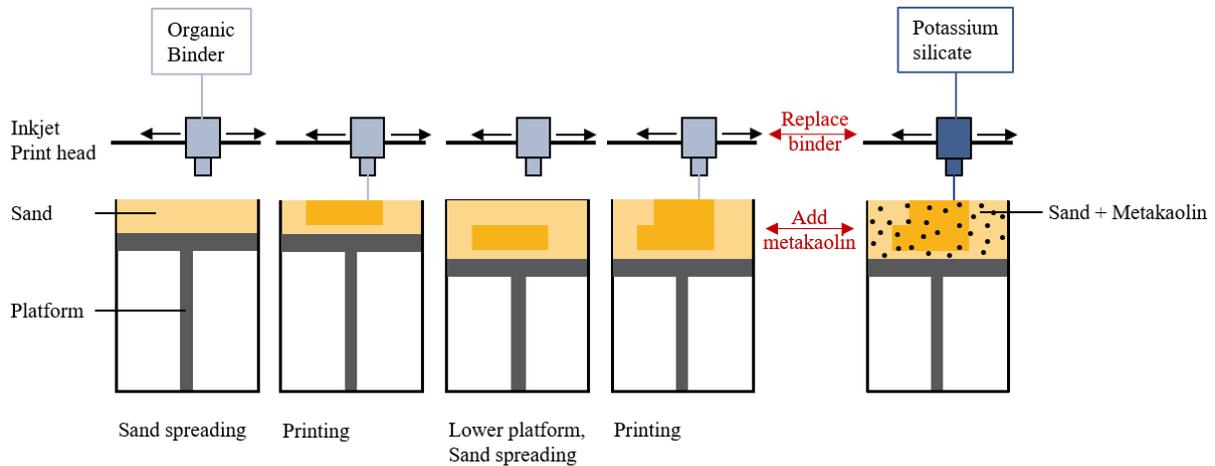


Figure 1: Left. Working principle of binder jet 3D printing. Right. Implementation of a geopolymer binder system. [8]

Therefore, powder bed binder jetting is a feasible construction technique, which helps to reduce the resource consumption. By using less material, additionally the CO₂ emissions are reduced. However, some alternatives to the current used materials need to be found. Indeed, commercially available binder jet 3D printing uses a powder bed of monosized silica sand glued with an organic binder. These organic binders are not suitable for indoor environments since they release volatile organic compounds (VOC), which are not healthy to breathe [9]. Additionally, the end of life scenario of the printed parts is not environmentally friendly [10].

In previous work, we showed that the usual system (organic binder on nonreactive powder bed) can be replaced by an inorganic solution on reactive powder, resulting in a strong binder between inert grains: a geopolymer [8]. Geopolymers are inorganic amorphous materials containing a covalently bonded alumina-silica network with sodium or potassium as charge compensators [11], often used in construction as cement replacements and fire resistant coatings [12]. Here, the implementation of geopolymers in binder jet 3D printing differs from the approach of Xia and co-authors where all the reactants are in the powder bed and are activated with water [13]. For better storability of the materials, the reactants are separated: the reactive aluminosilicate powder (metakaolin) is part of the powder bed and activated with an alkaline printing solution deposited at the surface (*cf.* Figure 1).

With the use of geopolymer instead of organic binder, the CO₂ emission of the printed product can be reduced significantly [10].

However, the process still contains natural sand, which is a scarce material [3]. Therefore, in this work a local waste material replaces silica sand in binder jet 3D printing. Instead of natural silica sand, waste stone material from a local gneiss quarry (Switzerland) is used. Using this material in 3D printing would promote a local waste material and could help to keep the business of small quarries profitable.

Two batches of samples were printed with quarry waste and sand respectively. The samples were compared in terms of compression strength and accuracy, allowing to state on their ability to be used in construction.

2. Materials and Methods

2.1. Mix design strategy for geopolymer binder jet 3D printing

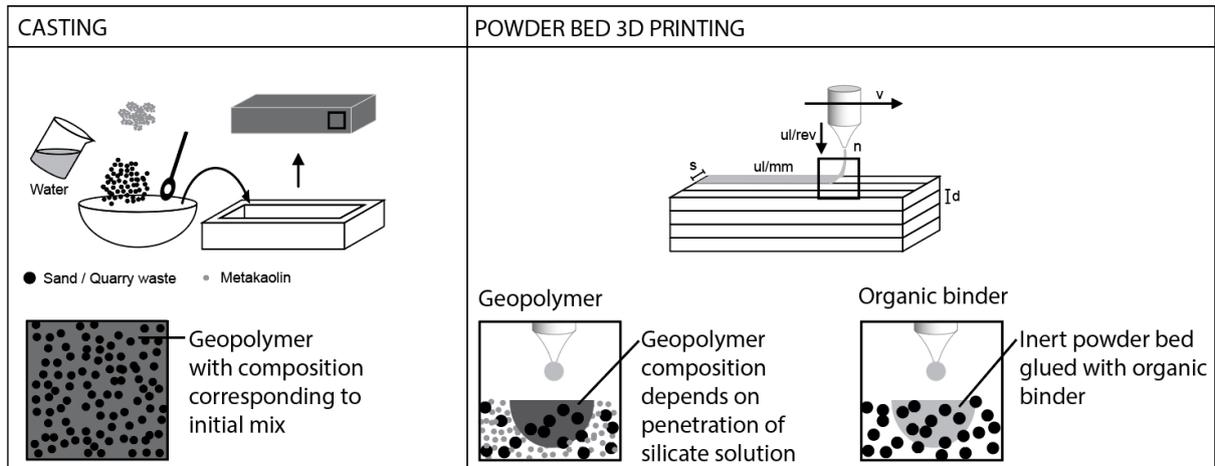


Figure 2: Graphical summary of difference in mix design between casting and 3D printing

Mix design for geopolymers in powder bed 3D printing differs from mix design of geopolymers for casting as well as the mix design of powder bed 3D printing with organic binder.

The difference to commercial binder jet 3D printing with organic binder is that the powder bed is not inert. The phenolic binder is enough to glue the sand, whereas our geopolymer approach is a two component glue with one component in the powder bed (metakaolin), and the other in the printing solution. The composition of the final geopolymer depends on the penetration of the liquid into the powder bed. This is as well the difference to geopolymer mix design for casting, where all the needed components can just be mixed.

In previous work, we showed that the composition of the alkaline printing solution can be determined from the compacity of the powder bed. Assuming the liquid to fill all the voids in the powder bed, the local liquid/solid ratio is known. From this, the silica, sodium, and water content of the solution can be tuned to obtain Si/Al, K/Al, H₂O/K₂O and SiO₂/K₂O ratios resulting in strong geopolymers.

2.2. Powder bed

In a stone quarry different types of stone waste accumulate. First, there is the fine sawing dust that is created when the stone is cut in plates. Second, not all the stone parts fulfil the quality requirements, and cannot be sold. In this work, the second sort of quarry waste is grinded to particle sizes below 1 mm and used as a replacement for silica sand (Quartz sand FS003 from Strobel Quartz Sand GmbH, mean grain size d_{50} of 240 μm) in powder bed 3D printing. The considered waste comes from a local gneiss quarry (Ticino, Switzerland) and consists of 45% plagioclase, 35% quartz, 10% chlorite, 5% zoisite, and 2% potassium feldspar (in volume). As a reactive powder Metakaolin (Metastar501 from Imerys, here referred to as MK) with a mean grain size d_{50} of 4 μm (measured with laser scattering, Horiba Partica LA-950) is added to the powder bed. The SiO₂ and Al₂O₃ contents of the Metakaolin are 51.63 wt%, and 44.37 wt%, respectively, and were determined with XRF.

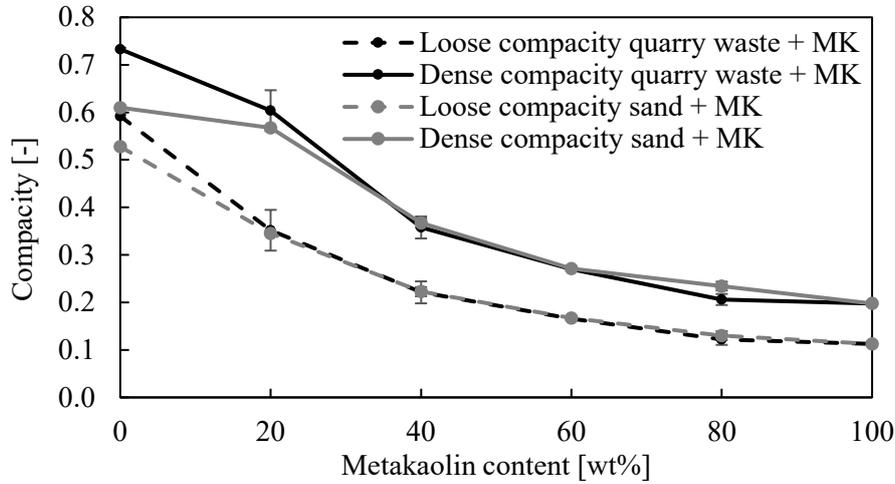


Figure 3: Loose and dense compacity of sand and quarry waste mixed with metakaolin

The loose and dense compacities of powder beds with varying amounts of MK were measured according to Lédée *et al.* [14]. The compacity of the pure quarry waste is higher than the compacity of pure sand due to the broader grain size distribution. As soon as MK is added to the mix, it dominates the behavior of the compacity, and the two mixes are similar (*cf.* Figure 3). Therefore, the same amount of MK was added to the quarry waste powder bed as in the previously printed reference with silica sand, namely 20 wt% [8].

2.3. Printing solution

Since the compacity of the quarry waste powder bed (with added 20 wt% MK) is equal to the compacity of the powder bed of the reference (silica sand + 20 wt% MK), the same potassium silicate solution was used as activating solution.

The solution was produced from deionized water at 20 °C, KOH in pellet form sourced from Sigma Aldrich, and silica gel in powder form (Davisil grade 635, Sigma Aldrich). The H₂O/K₂O and SiO₂/K₂O ratios were 13.9 and 1.6 respectively. The specific density of the solution (1.42 g/cm³) was measured with a Densito 30 PX (Mettler Toledo).

2.4. 3D printing

The used 3D printer is a self-built setup, designed with affordable and easily accessible components. The printer allows the use of different materials, and different parameters can be controlled: nozzle diameter (n), injection volume per distance ($V/dist$), spacing between printed lines (s), and the layer height (d). The nozzle diameter, the line spacing, and the layer height were set to 0.2 mm, 0.8 mm, and 2.5 mm, respectively. The injection volume per distance was varied between 0.5 μ l/mm, 0.75 μ l/mm, and 1 μ l/mm. The different printing parameters can be combined to the printed solution per volume of powder bed:

$$\frac{\text{solution } [\mu\text{l}]}{\text{powder bed volume } [\text{mm}^3]} = \frac{V/dist [\mu\text{l}/\text{mm}]}{n [\text{mm}] * s [\%] * d [\text{mm}]} \quad (1)$$

Three cubes with a targeted size of 4 cm x 4 cm x 4 cm were printed of each variation. After 7 days the samples were tested on compression in a mechanical press (Unitronic from Matest) using a 50 kN cell, applying 0.1 kN/s. The dimensions of all the cubes were measured in x, y, and z direction and the accuracy was determined as followed:

$$\text{Accuracy [\%]} = \frac{\text{measured size [mm]} - \text{targeted size [mm]}}{\text{measured size [mm]}} * 100 \quad (2)$$

2.5. Casting

For comparison, geopolymer samples with sand and quarry waste were casted with a liquid to solid ratio of 0.4. This ratio corresponds to the local ratio in the printed objects, estimated from the compacity of the powder bed. The powder mixes (20 wt% MK) were mixed with the potassium silicate solution for 2 minutes in a mechanical mixer (Auto-mortar mixer from Matest) and cured at room temperature for 7 days.

3. Results

3.1. Compression strength

All the printed samples show compression strengths between 10 and 12 MPa (*cf.* Figure 4), independent of their powder bed and amount of liquid per volume. The geopolymer prints are lower in strength than their corresponding cast (37 MPa for quarry waste and 42 MPa for silica sand), but twice as strong as a print with silica sand and phenolic binder (5.5 MPa, printed in a commercial printer [5]).

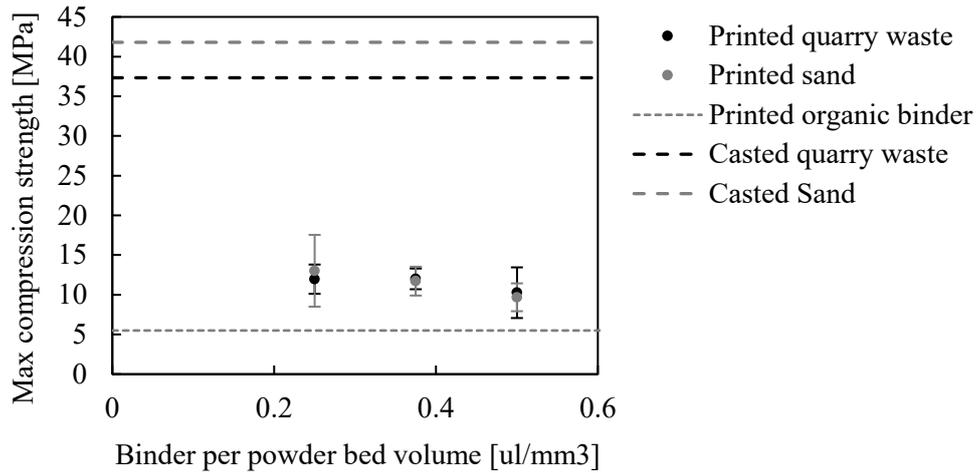


Figure 4: Compression strength of 3D printed and casted samples

3.2. Precision and accuracy

For all the printed samples the precision in z direction is the highest (smallest standard deviation), closely followed by x and y (*cf.* Table 1). The accuracy calculated with eq. 2 is very similar in all directions (*cf.* Table 2). The samples printed with the quarry waste are as precise and accurate as the reference printed with sand. There is no remarkable difference in precision or accuracy between the samples printed with different amounts of liquid per distance.

Table 1: Dimensions with standard deviation of printed samples

V/dist ul/mm	Quarry waste			Silica sand		
	x mm	y mm	z mm	x mm	y mm	z mm
0.5	43 ± 0.8	42 ± 0.5	43 ± 0.0	42 ± 0.3	42 ± 0.6	43 ± 0.4
0.75	42 ± 0.5	42 ± 1.0	42 ± 0.3	42 ± 0.7	42 ± 0.9	43 ± 0.4
1	42 ± 0.9	42 ± 1.1	42 ± 0.3	42 ± 0.7	42 ± 0.6	43 ± 0.0

Table 2: Accuracy of printed samples, calculated with eq. 2

V/dist μl/mm	Quarry waste			Silica sand		
	x %	y %	z %	x %	y %	z %
0.5	6.6	5.3	6.2	5.0	4.2	6.9
0.75	6.1	5.9	6.0	5.8	5.4	6.7
1	5.3	5.6	5.7	4.8	4.4	7.5

4. Discussion

The printed samples show lower compression strengths than the casted, but there is no loss in strength using quarry waste instead of silica sand.

The lower strength of the printed parts can be explained by the lower overall density. Locally the same geopolymer is built in the print and in the cast, but the printed material still contains unreacted and unglued powder, leading to lower strengths.

Parts displaying 10 to 12 MPa are strong enough to be used in compression-only structures [15], or as lost formworks for complex concrete parts [5]. The high porosity of the parts does not allow an outdoor application due to the risk of freeze-thaw. However, the high porosity is beneficial for the hygroscopic properties. De Rossi and co-authors [16] showed that geopolymers built from waste have good hygroscopic properties. Further tests on the controllability of the moisture buffering capacities by varying printing parameters have to be done. This would provide an additional advantage compared to the commonly used organic binder, which is not suitable for indoor applications due to the released volatile organic compounds.

From experiments on a previous edition of the printer with higher layer thicknesses, it was expected that increasing liquid per distance lead to increased compression strengths and decreased accuracy. This is not visible in the tested range. The compression strength as well as the accuracy are constant throughout the measured samples. To optimize the amount of printed liquid, final compression strength, and accuracy, more samples with lower and higher $\mu\text{l}/\text{mm}^3$ will be printed and tested.

Powder bed 3D printing seems to be an ideal application for the gneiss quarry waste. Samples printed with the quarry waste instead of silica sand show the same properties in terms of compression strength and accuracy.

The natural stone industry is an important activity in the economies of the valleys in Ticino, but the sector is in a crisis. Especially the high prices to deal with the waste material puts pressure on the finances of the small quarries. The waste pieces could be used as aggregates in concrete, but unfortunately this is not economical anymore. Nowadays it is cheaper to import aggregates with higher quality from Italy. Currently, there is no application for the waste from the quarries and it needs to be landfilled. Since local space is very limited, the stone is exported to Italy, and landfilled there. The use of the grinded quarry waste in 3D printing could help to provide new markets to the quarries. They could serve the high demand of 3D printed objects with their local material, and at the same time they could save money by not disposing and transporting the material.

The next step would be to replace also the reactive powder (MK) with quarry waste. Here for, the fine waste from sawing could be used, activated by alkali fusion [17]. This will be investigated in further work.

5. Conclusion

With powder bed 3D printing structurally optimized building parts and formworks can be printed, which allows a reduced material consumption. If the printed materials are recycled and waste materials, additionally the CO_2 emissions can be reduced.

Previously it was shown that the organic glue in powder bed 3D printing can be replaced by an inorganic geopolymer glue. In this work, we successfully replaced the silica sand by waste granulates from a local quarry in the south of Switzerland. The use of this quarry waste does not only help to minimize the use of the scarce sand, it also helps small quarries to keep up their business.

4 cm x 4 cm x 4 cm test cubes were printed on a self-built printer with different amounts of printing solution per volume of powder bed. All the samples printed with quarry waste reach the same compression strengths, accuracies and precisions as the reference printed with silica sand. Therefore, we could show that the scarce silica sand used in powder bed 3D printing can be replaced with the local waste material from a gneiss quarry.

To use 3D printed quarry waste for printing structurally optimized building parts, more complex shapes than rectangular bars are needed. By printing a curved piece with holes and openings, it was shown that this is not an issue for the present set up (*cf.* Figure 5). Currently the maximum printing size is about 50 cm x 40 cm x 35 cm but technically it is possible to build a bigger printer with a larger sample size. To increase the printing speed, additional print heads could be added. The only thing that changes from a material point of view when going to bigger scale, is the setting time between layers. Since final setting only takes place after 14 hours, the layers will still bound to each other, even if the printing time per layer is longer for bigger parts. Thus, this technology has a high potential to be scaled up and used in construction.

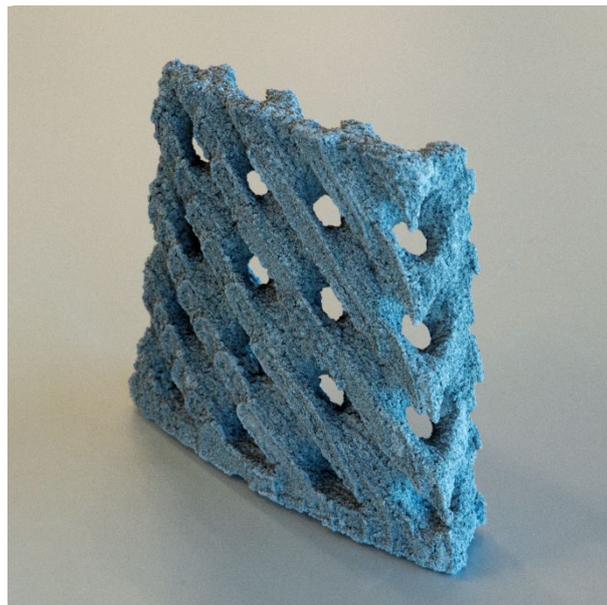


Figure 5: Complex shape printed with quarry waste

Acknowledgements

The Swiss National Fund (SNF, project N°200020_169359) and SUPSI (project 12RAY1CAST3D) are gratefully acknowledged for their financial support.

References

- [1] A. Horvath, "Construction materials and the environment," *Annu. Rev. Environ. Resour.*, vol. 29, pp. 181-204, 2004.
- [2] D. Ioannidou, G. Meylan, G. Sonnemann, and G. Habert, "Is gravel becoming scarce? Evaluating the local criticality of construction aggregates," *Resources, Conservation and Recycling*, vol. 126, pp. 25-33, 2017.
- [3] P. Peduzzi, "Sand, rarer than one thinks," *Environmental Development*, vol. 11, pp. 208-218, 2014.
- [4] A. Jipa, M. Bernhard, B. Dillenburger, and M. Meibodi, "3D-Printed Stay-in-Place Formwork for Topologically Optimized Concrete Slabs," in *2016 TxA Emerging Design+ Technology conference*, 2016: Texas Society of Architects.
- [5] M. Rippmann, A. Liew, T. Van Mele, and P. Block, "Design, fabrication and testing of discrete 3D sand-printed floor prototypes," *Materials Today Communications*, vol. 15, pp. 254-259, 2018.
- [6] R. A. Buswell, W. L. de Silva, S. Jones, and J. Dirrenberger, "3D printing using concrete extrusion: a roadmap for research," *Cement and Concrete Research*, 2018.
- [7] D. Lowke, E. Dini, A. Perrot, D. Weger, C. Gehlen, and B. Dillenburger, "Particle-bed 3D printing in concrete construction—possibilities and challenges," *Cement and Concrete Research*, vol. 112, pp. 50-65, 2018.
- [8] V. Voney *et al.*, "Powder Bed 3D Printing with Geopolymers," presented at the 2nd RILEM Spring Convention & International Conference on Sustainable Materials, Systems and Structures (SMSS2019), Rovinj, Croatia, 2019.
- [9] N. Bakhiya and K. E. Appel, "Toxicity and carcinogenicity of furan in human diet," *Archives of toxicology*, vol. 84, no. 7, pp. 563-578, 2010.
- [10] I. Agustí-Juan, A. Jipa, and G. Habert, "Environmental assessment of multi-functional building elements constructed with digital fabrication techniques," *The International Journal of Life Cycle Assessment*, pp. 1-13, 2018.
- [11] J. L. Provis and J. S. J. Van Deventer, *Geopolymers: structures, processing, properties and industrial applications*. Elsevier, 2009.
- [12] P. Duxson, A. Fernández-Jiménez, J. L. Provis, G. C. Lukey, A. Palomo, and J. S. van Deventer, "Geopolymer technology: the current state of the art," *Journal of materials science*, vol. 42, no. 9, pp. 2917-2933, 2007.
- [13] M. Xia and J. Sanjayan, "Method of formulating geopolymer for 3D printing for construction applications," *Materials & Design*, vol. 110, pp. 382-390, 2016.
- [14] V. Lédée, F. De Larrard, T. Sedran, and F. Brochu, "Essai de compacité des fractions granulaires à la table à secousses," *Laboratoire Central des Ponts et Chaussée—IST Diffusion des éditions, Juillet 2004, Paris*, 2004.
- [15] M. Rippmann and P. Block, "Digital Stereotomy: Voussoir geometry for freeform masonry-like vaults informed by structural and fabrication constraints," in *Proceedings of the IABSE-IASS Symposium*, 2011, vol. 2011.
- [16] A. De Rossi *et al.*, "Waste-based geopolymeric mortars with very high moisture buffering capacity," *Construction and Building Materials*, vol. 191, pp. 39-46, 2018.
- [17] L. Tchadjé *et al.*, "Potential of using granite waste as raw material for geopolymer synthesis," *Ceramics International*, vol. 42, no. 2, pp. 3046-3055, 2016.