63. Development of improved bricks (LM) and use of new technologies for ecological bricks (LE) elaboration

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Abstract Brick is a material that has been widely used in construction sector, however, it has not received the necessary attention in order to improve its performance, which has resulted a general lack of knowledge about its manufacturing processes such as: clay mines over-exploitation, ignorance of mixtures dosages and greenhouse gas emissions from wood burning.

This research therefore aims to find a suitable mixture to produce improved bricks (LM) by analysing its mechanical properties compared with handmade bricks (LPA). Besides, this research work uses the "geo-polymerization" process for the preparation of ecological bricks (LE) at slightly high temperatures which gives a significant environmental benefit.

In order to achieve the objectives, firstly, samples were characterized from mines by physical, chemical and mineralogical tests and then, it was determined the optimal mixture for LM and LE elaboration by evaluating its optimal factor (OF) according to the diametric compression test. In order to perform this test, specimens of different combination of materials and processes were elaborated. Once optimal mixtures were obtained, bricks were produced to measure their mechanical resistance by the performance of simple mechanical compression test.

Results show that LM resistance is increased by 400% over handmade bricks from the study area. Moreover, the use of waste from mines as raw material for the elaboration of LE shows that their resistance value exceeds 150% of stablished in regulations.

Keywords: Clay bricks; Mixtures optimization; Geo-polymerization; Alkaline Activation; Simple mechanical compaction.

1 Introduction

Brick is the oldest building construction material worldwide used by humanity (Bianucci 2009). Generally, bricks are handmade manufactured from non-renewable resources (such as clay) and have low production cost. Bricks offer several properties such as durability, strength, resistance to climate changes, temperature, sound insulation, high fire resistance and thermal transmittance (García, et al, 2016).

Nowadays, there is a general ignorance of its composition: microstructure and mechanical performance. Some researchers are encouraged to seek new raw materials, alternatives to clay, for bricks manufacture and production in order to minimize over-exploitation of clay mines and environmental pollution since the actual raw material used requires high temperatures for cooking (Chen, et al, 2011; Roy S et al 2007). Romero and Flores (2010) used waste from mineral extraction to solve this environmental problem and also to directly improve the added value to this type of waste.

The southern region of Ecuador is not exempt from this problem, which is evident due to the intensive exploitation of clay mines. Whereas, this research aims to optimize handmade bricks mixtures (LPA) for the manufacture of improved bricks (LM). Besides, it is also studied the possibility of reusing mines waste for the production of ecologic bricks (EC) according to their chemical characteristics.

For the preparation of LM, the silica of the raw material is transformed into cristoballite by the sintering process of temperature of 950 ºC. In the case of LE, it is applied the geo-polymerization method where the alkaline solution dissolves the contents of silica and alumina, forming a geo-polymeric gel at a temperature below 120 ° C.

2 Materials and Methods

2.1 Study area localization

The study is performed in southern Ecuador. For the preparation of LM, mines were selected according to their availability and accessibility and collecting samples from five sectors: Ceibopamba (MCB), Cangahua (MCA), Chinguilamaca (MCH), Palanda (MPL) and fine sand from the Chinguilamaca (MAF) sector, located in the parish of Malacatos which belongs to the province of Loja.

Ecological bricks raw material was extracted from mine tailings (RM), which are produced by the gravity concentration process and do not generate heavy metal pollution, thereby selecting the granting “Minera Pituca II” COD 500648, Timbara parish of the province of Zamora Chinchipe.

2.2 Raw material characterization

2.2.1 Physical tests

For the soil classification, a particle size analysis is performed, applying ASTM standard D422 (hydrometer method) and ASTM standard D4318 that determinates the Atterberg limits: liquid limit (LL), plastic limit (PL) and plastic index (PI).

2.2.2 Chemical tests

Though these tests, it was obtained the carbonate content, organic matter, hydrogen potential level (pH) and the specific surface. Chemical composition was obtained by the performance of the X-rays fluorescence test (XRF). The thermogravity analysis (TGA) was only applied in the preparation of Improved Bricks.

Each sample experimented variations between ambient temperature and 1000 ºC at a heating rate of 20 ºC/minute. The thermal analysis shows the curve of the variation of temperature importance (TGA, green): the first derivative of the temperature (DSC, blue) represents the gaining speed or the weight loss and the second derivative is the heat flow (2D-DSC, brown).

2.2.3 Mineralogical tests

X-Rays Diffraction: This method determines the mineralogy of raw material, not only identifying its origin possibilities, but also to allow to study its crystallographic characteristics. For the analysis results, it was used the DIFFRAC.SUITE Bruker EVA software and it was registered a step of 0,02 degrees and a measurement time of 2 seconds for each sample.

2.2.4 Diametric compression (Brazilian test)

This test is an indicator of the optimal mixture for different combinations of materials thus optimizing time and resources. It requires the utilization of a load cell named LC model 101-5K and a press called Versa Tester, at a loading rate of 0,2 mm/min.

The compression resistance value of items is calculated by the following formula:

$$\sigma_{CD} = \frac{2P_{máx}}{\pi De} \quad (1)$$

Where "Pmax" represents the maximum resistance value of mixtures, "D" is the tested specimen diameter and "e" is the thickness of the specimen.

For mixtures optimization, it was applied an optimization factor (OF) according to the described process by Del Coz Díaz, et al (2001). Thus, OF is defined as the quotient between the maximum diametric compression resistance value which is expressed in MPa and mixture weight P, expressed in kg "Ec. 2".

$$FO = \frac{\sigma_{CD}}{P} \qquad (2)$$

Optimal mixtures present the highest value of the optimization factor, OF.

This test was performed to the specimens that were prepared with different mixtures of clays and cooked to 950 °C (**Fig. 1**), as well as those made with combinations of process of variables of geo-polymerization and dried at a temperature lower than 200 ° C (**Fig. 2**).

3 Experimental design

3.1 Improved bricks (LM)

Currently, producers of bricks from the study area apply ancestral concepts, obtaining a combination of "clay material" and "sand material" in proportions of 90/10 respectively. However, in order to obtain improved mixtures, OF was used, for which six types of combinations were performed in different percentages (**Fig. 1**)

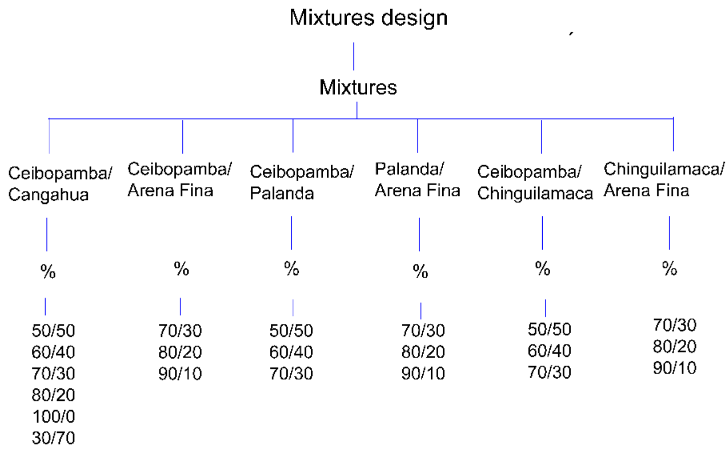

Fig. 1 Mixtures design for handmade bricks. **Source:** Authors' own.

Materials were carefully handmade mixed. Each mixture rested for a period of 12 hours in sealed bags to perform a correct maturation process and consequently, achieve the maximum plastic level. After this, portions of each mixture were placed into moulds of 75 mm of diameter and 20 mm of thickness. Three replicates of each mixture were prepared and it was also placed a 4 kg weight during 2 hours for the compaction. After, specimens were dried until they reached 105 ºC and cooked to 950 ºC.

Though OF use, 6 mixtures were selected and specimens elaborated which were sintered at 500, 700 ad 950 ºC to determine the optimal cooking temperature.

Finally, these six optimal mixtures were used to prepare Improved Bricks of dimensions of 28 x 14 x 9 cm, which were analysed by the simple compression test. These test results were compared to the Ecuadorian standard, NTE INEN 297 and the LPA.

3.2 Ecological bricks (LE)

For the determination of the optimal mixture for LE elaboration, it was used combinations of different variables that perform during the chemical process such as: solution content, dry temperature and moulding pressure. After, raw material was characterized and the alkaline activator selected. The activator that provided the best physical and mechanical characteristics was the solution of sodium hydroxide and was used as a precursor in the geo-polymerization process.

For the specimens' elaboration, different combinations of alkaline solution were applied in different molarity concentrations with raw material in various percentages in solution content (**Fig. 2**). Specimens were introduced in the stove for 72 hours to allow the alkaline solution to act on the raw material. After, they were dried at temperatures of 90, 120, 150 and 180 ºC and finally, it was performed the diametric compression test to select the optimal mixture by OF application.

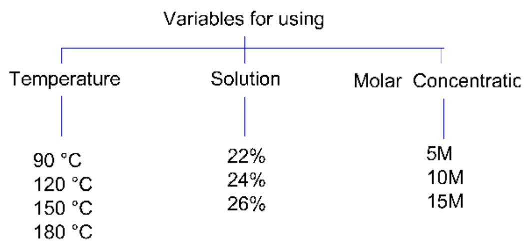

Fig. 2 Variables used for the geo-polymerization process. **Source:** Authors' own.

4 Results analysis

4.1 Raw material characterization

Physical test results show the presence of clay in soils from the mines of Ceibopamba and Cangahua and fine sand in the provided from Chinguilamaca and Palanda mines. Materials from "La Pituca II" tailings were classified as sandy and mud soils.

The chemical composition of LM and LE materials is shown in **Fig. 3**, where it is observed that the most existence chemical component is silicon oxide after aluminium oxide, both principal's alumina-silicates compounds.

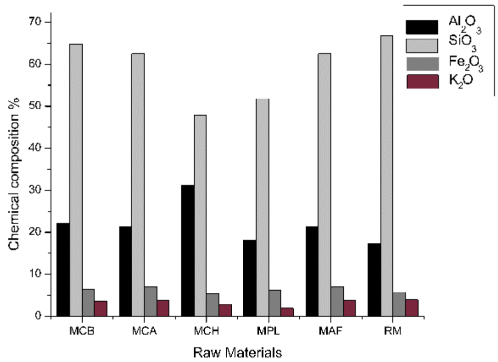

Fig. 3 Chemical composition. FRX. **Source:** Authors' own.

In Table 1, it is shown the chemical composition of used raw materials: Hydrogen potential that indicates the alkalinity of mixtures, specific surfaces that relates the existence between area particles and mass.

Mineralogical DRX test reveals that mixtures for the LM elaboration presents quartz as dominant mineral, which helps the improvement and maintenance of durability after removing mixtures from moulds (Quillupangui, 2011). After this main material, it is found the kaolinite which adds refractory characteristics to bricks and improve their mechanical properties.

Results of the components for the LE elaboration reveal the presence of mineral of low quart, illite and chlorite, therefore mine waste are generated by high amounts of silica. Results are corroborated within the X-Rays fluorescence test.

In **Fig. 4**, it is presented the diffractogram for mine tailings mixtures and Ceibopamba.

Table 1. Mineralogical and mechanical tests

Samples	Chemical tests		Mineralogical tests
	pH	Specific surface m^2/g	DRX
Ceibopamba	7.7	6.1	Quartz, Kaolinite, Hematite, Montmorillonite, Illite
Cangahua	7.2	48.9	Quartz, Kaolinite, "Moscovita" and Hematite.
Chinguilamaca	5.9	90.5	Quartz, Hematite, Montmorillonite, Illite
Palanda	8.6	20.2	Quartz, Kaolinite, Hematite, Illite
Fine sand	7.2	39.8	Quartz, Kaolinite "Moscovita", Illite
Mine tailings	8.6	10.4	Low Quartz, Illite, Chlorite, Hematite, "Oligoclasa"

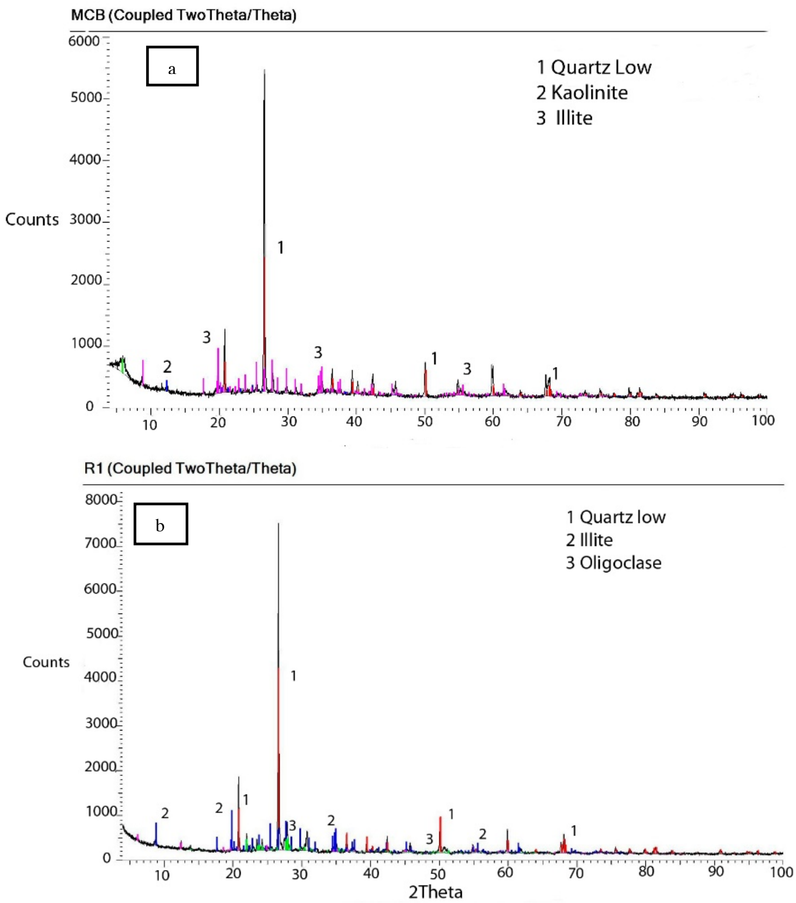

Fig. 4 X-Rays diffractogram: a) MCB: Ceibopamba, b) R1: Mine tailings. **Source:** Authors' own.

Fig. 5 shows the analysis of the TGA curve for the Ceibopamba mixtures. Firstly, it is observed the endotherm process due to initial heating reaching to 120 ºC, losing 2,202% of its mass. The DSC curve value of 75 ºC indicates a mass loss. Furthermore, during the section between 120 and 170 ºC, TGA curve decreases by 0,297 % due to hygroscopic illite water loss.

Arsenović, et al (2014) defines that the oxidation of organic matter is achieved between temperatures of 200 and 500 ºC, at temperatures of 310 ºC and 610 ºC, it is presented the most significant mass loss, 2,245%. The DSC curve top value at 490 ºC shows the decrease of mass at this section. The illite mineral dihydroxylation is manifested at 573,43 ºC (D2-DSC curve)

Linares, et al (2013) establishes that metakaolinite is the kaolinite evolution and because it its raised to 950 ºC, silica becomes cristobalite and a spinel structure is formed. Thermograms recorded for mixtures from Cangahua, Chinguilamaca, Palanda and fine sand show similar thermal behaviours.

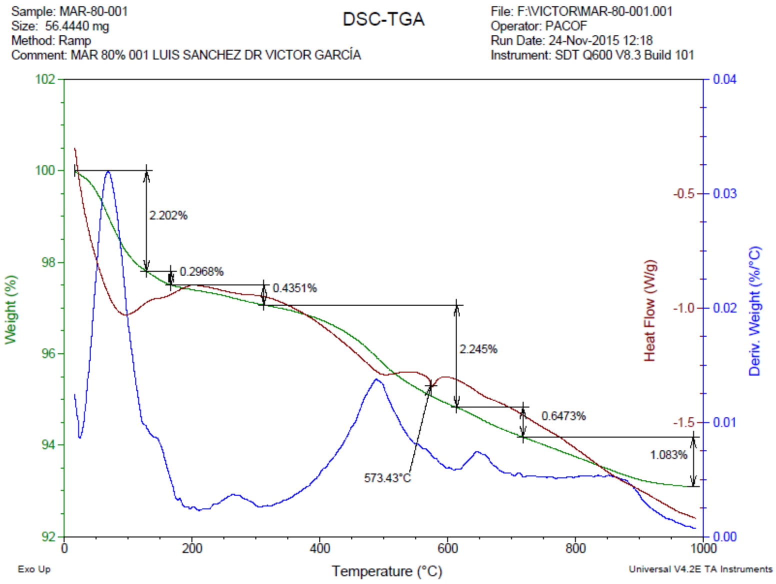

Fig. 5 Ceibopamba mixture thermograms. **Source:** Authors' own

4.2 Diametric compression test results

4.2.1 Improved bricks (LM)

Due to mixtures combinations variety and their diverse dosages percentages, it is necessary to take as an indicator the best OF values for mixtures optimization. From each combination, it was selected the best OF value, expect for the mixture between Palanda mine and fine sand that presented lower values.

Table 2 shows the Ceibopamba and Cangahua combination, determining the optimal mixtures in percentages of 60/40 and 80/20, showing the best result of diametric compression test (ft), lower contraction after drying (Css) and total contraction.

Table 2. Diametric compression (ft) y contraction (CT) test results for mixture: Ceibopamba (MCB) and Cangahua (MCA). (Del Coz Díaz, et al 2011)

Mixture	Percentage (%)	ft (Mpa)	FO (MPa/kg)	Css (%)	CT (%)
MCB/MCA	50/50	3,91	36,61	8,88	9,67
	60/40	4,49	44,79	3,31	**6,75**
	70/30	2,60	26,77	3,51	11,28
	80/20	4,72	47,00	3,25	**5,51**
	100/0	3,50	34,72	6,66	10,30
	30/70	3,52	34,53	2,18	7,01

Furthermore, OF indicates the higher values for mixtures of 60/40 and 80/20 percentages, which can be observed in **Fig. 6**.

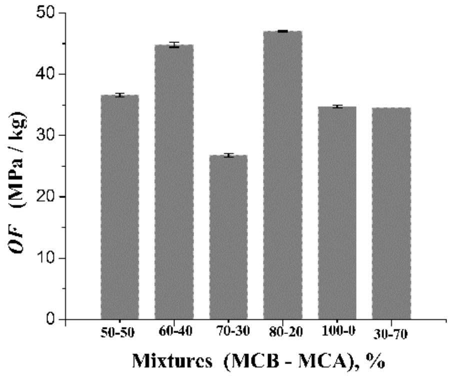

Fig. 6 OF of Ceibopamba and Cangahua mixtures. **Source:** Authors' own

Similarly, this methodology was performed with all combinations that are established in **Fig. 1** and a total of six optimal mixtures was obtained and are presented in Table 3.

Table 3 Best optimal factor (OF) mixtures.

ID	SAMPLE 1	SAMPLE 2	Percentage (%)	F.O Mpa/Kg
1	Ceibopamba Mine	Cangahua Mine	80/20	47,0
2	Ceibopamba Mine	Cangahua Mine	60/40	44,79
3	Ceibopamba Mine	Fine Sand Mine	90/10	20,7
4	Ceibopamba Mine	Palanda Mine	70/30	16,45
5	Ceibopamba Mine	Chinguilamaca Mine	70/30	22,37
6	Chinguilamaca Mine	Fine Sand Mine	90/10	17,17

In order to obtain a curve of the variation of the diametric compression effort versus the cooking temperature (**Fig. 7**), it was prepared specimens of best OF values such as ID 1, 2, 3 (Table 3), concluding that the best cooking temperature is 950 ° C.

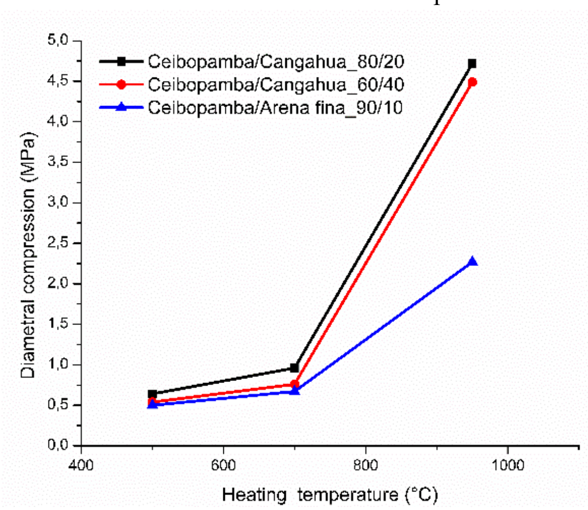

Fig. 7 Variation of the diametric compression effort according to temperature. **Source:** Authors' own

4.2.2 Ecological bricks (LE)

The first variable analysed through specimens was temperature while constantly maintaining the solution content (SC) and molar concentration (MC). It was obtained an increase of the OF value, from 3.29 to 8.25 MPa/kg and temperatures of 90 and 180 ºC respectively. Results show that temperature is beneficial is these conditions (Table 4).

Table 4 Diametric compression results (constant CS and CM) for different temperatures.

CS (%)	CM (M)	T (°C)	D (cm)	E (cm)	Charge (kg)	σt (MPa)	FO (MPa/kg)
22	10	90	5.04	1.65	22.60	0.169	**3.29**
		120	5.05	1.81	26.17	0.180	3.68
		150	5.04	1.54	42.80	0.342	6.44
		180	5.07	1.62	58.76	0.439	**8.25**

However, if solution content increases, different results are obtained (Table 5). It is observed a significant decrease of OF values when solution content changes from 204% to 26%. This values decrease is due to the rapid evaporation of the solution without the chemical process development.

Table 5 CS variation and its effect in diametric compression test.

CS (%)	CM (M)	T (°C)	D (cm)	e (cm)	Charge (kg)	σt (Mpa)	FO (MPa/kg)
24	10	90	5.07	1.55	27.84	0.220	4.31
		120	5.03	1.55	86.39	0.691	**14.99**
		150	5.09	1.52	42.14	0.336	6.46
		180	5.15	1.58	40.20	0.315	6.08
26	10	90	5.11	1.36	23.41	0.211	0.21
		120	5.11	1.45	69.38	0.587	0.59
		150	5.03	1.49	28.77	0.241	**4.84**
		180	5.20	1.51	26.85	0.211	4.2

Finally, it is related the molar concentration (MC) and solution content (SC) for different temperatures, obtaining a resistance of 5 MPa/kg for a MC of 5 and a SC of 22%. However if it is only varied the SC to 26% at a 90ºC temperature, resistance value is three times incremented.

Molar concentration confers good mechanical properties, obtaining results rear to 21 MPa/kg at temperatures of 120 ºC and a SC of 26% and MC values of 15. It is important to indicate that an increase of the solution causes an alteration in the geo-polymerization process due to excessive saturation of raw material without reaching the aluminium-silicate gel precipitation.

4.2 Simple compression test results

Simple compression test results are presented in **Fig. 8a** for both LPA and LM and are compared to Ecuadorian legislation. LPA show maximum resistance values of 5 MPa, not complying the standards (min 8 MPa). Due to mixture optimization, LM resistance increases by 400%, reaching values of 20 MPa. This resistance increase is due to the spinel structure transformation which is produced by the transformation of silica into cristoballite since raw material temperature is elevated to 950 ºC.

Bricks elaborated by the geo-polymerization process with an alkaline solution of 15M and solution content of 26 %, present better compression resistance results obtaining values of 12.1 MPa, which indicates that the chemical process formed a stable structure due to silica-aluminium components dissolution and their respective hardening due to the poly-condensation stage. Furthermore, temperature is an important factor that affects the chemical process and constituently, the mechanical aspect of bricks. For mine waste used in this research, optimal mature temperature is 120 ºC.

Therefore, if the selection of the items elaboration conditions is adequate, ecological bricks may be elaborated by adding mine waste, complying with standards.

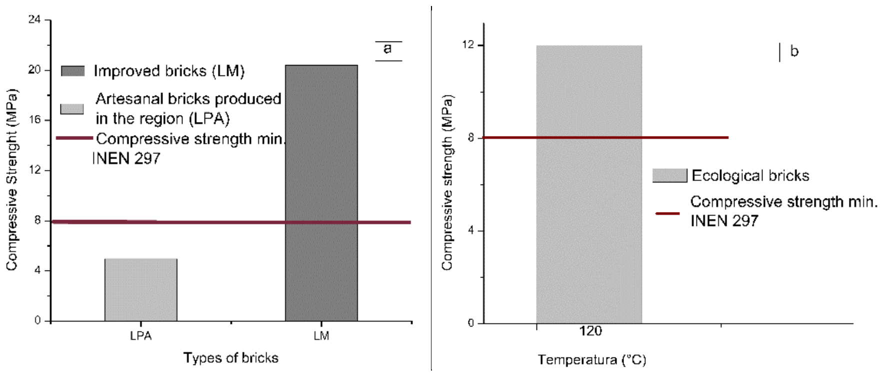

Fig. 8 a) Simple compression results vs. thermal treatment. b) Simple compression results of different brick types. **Source:** Authors' own

5 Conclusions

Simple compression test results show a notable resistance of Improved Bricks. It was obtained an increase of 400% compared to Handmade Bricks in compliance with Ecuadorian regulations. It was achieved through the use of the optimal mixtures obtained from Ceibopamba and Cangahua mines of percentages of 80/20 respectively. Therefore, environmental impact is reduced, derivate from an intense exploitation of clay mines due to a general ignorance of mixtures dosages.

Mine residues are suitable for Ecological Bricks elaboration, due to their acceptable chemical and mechanical characteristics to improve the geo-polymerization process. Optimal values present the same composition: solution content of 26 %, alkaline concentration of 15M and temperature of 120 ºC. These provide mechanical characteristics that are ideal for their use in buildings construction, exceeding the minimum values that are suggested in Ecuadorian regulations by 150%. Mine tailings use in bricks elaboration is technically possible and an alternative to clay use, minimizing the environmental impact generated by bricks cooking process due to wood burning at high temperatures.

6 References

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Acknowledgments

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