The use of Piasentina Stone for more sustainable cement

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Synopsis: This research focuses on the use of waste materials from other production processes available within the national territory, obtaining the dual advantage of effectively reducing the CO_2 released, but also of reusing substances that would otherwise have to be sent to landfill. In particular, the study concerns Piasentina Stone, which is a stone available in the Italian region of Friuli Venezia Giulia and which is used for flooring and decorative elements. To obtain the finished material ready for sale, the manufacturing process is characterized by more than 50% of waste products (large stone elements, crushed stone and cutting mud). The composition of the stone is characterized by a percentage of limestone greater than 95%, which makes it perfectly suitable both as a material to be used in the kilns for the production of clinker, and as a substitute for clinker in binary and composite cements. A campaign of chemical-physical investigations was conducted on the stone to evaluate its suitability with respect to the Codes limitations. Analyses on mortars obtained with partial replacement of clinker have been developed. The purpose of this research is to provide a possible alternative to obtain a more sustainable cement with a natural by-product widely available.

Keywords: Piasentina Stone, sustainability, mortar, CO_2 emissions

INTRODUCTION

Carbon dioxide emissions from cement production alone account for over 6% of global emissions, making it one of the most polluting sectors [1]. In particular, the most polluting phase is the clinker production phase: this is why innovative mixtures such as binary and ternary cements have become increasingly popular in recent years [2] [3]. These cements contain limestone, silica fume, blast furnace slag, pozzolana, fly ash and calcined shale, which are waste materials from other production processes, but which are often not available locally but must be transported by road over long distances. In this way, if we want to carry out a balance of CO_2 emissions, almost all the savings resulting from the reduction of clinker are offset by high releases of carbon dioxide related to transport, nullifying the beneficial effect. A more virtuous approach, which should be pursued by all cement producers, must be to exploit waste materials available directly in the local area, optimizing CO_2 emissions. This study provides an example of this correct way of working, focusing on the use of waste from the processing and extraction of Piasentina Stone, which is a limestone rock available in Friuli Venezia Giulia [4].

RESEARCH SIGNIFICANCE

Piasentina Stone is a widely available material in Friuli Venezia Giulia that is used for the construction of flooring and cladding; however, during its processing, scraps and waste are produced for more than 50% in weight. This material, currently classified as a by-product, can be exploited by local cement factories in their production chain, reducing the consumption of virgin resources and ensuring a certain percentage of recycled material. The aim of this study is to show the results obtained from tests on mortars with a partial replacement of clinker with Piasentina Stone, but above all to provide an example of a virtuous approach that can be reproduced in all areas of Italy with other locally available materials [5].

CO2 EMISSIONS OF CLINKER

The main cause of the high carbon dioxide emissions of cement is clinker, in fact for its production it is necessary to bring the limestone rock to high temperatures in order to develop chemical reactions of calcination. To convert the construction world towards more sustainable practices, it is essential to reduce not only the energy needed to produce the material, but rather the use of clinker within the binder mixtures, using alternative materials, which guarantee equivalent properties. Table 1 shows the CO_2 emissions related to some recurring materials [6] [7]:

Table 1–Emission of CO_2 of some recurring materials

Concrete components	CO_2 (kg CO_2/m^3)
Clinker	2.191
Fly Ash	9,6
Blast-Furnace Slag	160,8
Silica fume	33,6
Sand	12
Gravel	12

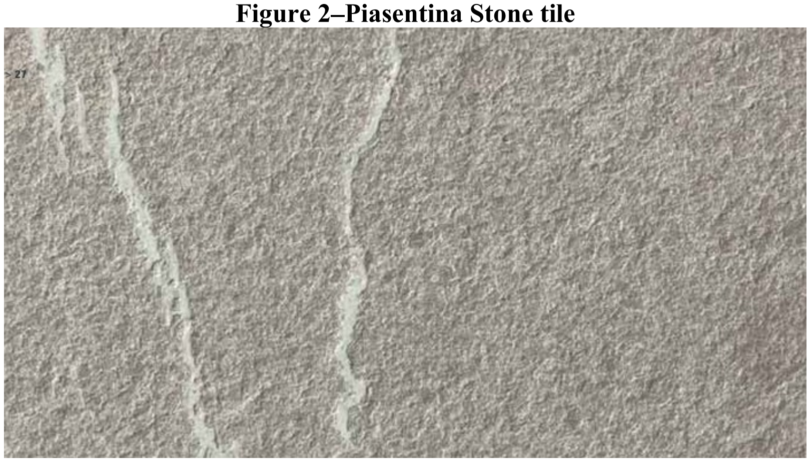
It can be seen that clinker has an emission at least two orders of magnitude higher than other materials: the use of substances such as Piasentina stone (or more generally limestone), which can achieve strong reductions in terms of emissions, which can lead to a beneficial effect in terms of environmental sustainability.

PIASENTINA STONE

Piasentina stone, geologically, is a calcareous sedimentary rock of secondary origin, deriving from the re-cementation of carbonate deposits; it is very common in the areas of north-eastern Italy on the border with Slovenia, in the Friuli Venezia Giulia region. The material is part of the carbonate megabanks that constitute the powerful succession of the Grivò Flysch. Petrographically, Piasentina Stone is identified as calcarenite. Commercially, it is used for the production of high-quality flooring for interiors and exteriors, with various surface treatments of pleasant color. During the cutting and finishing process of the material, different types of waste are produced, which account for over 50% of the virgin material extracted from the quarry: most of the volume is represented by the initial roughing, since the single block must be brought back to a linear and regular shape. Secondly, cutting waste is obtained, since slabs and parallelepeds of standard dimensions must be obtained, eliminating the excess compared to the starting block. Finally, a final product is mud: the cutting process of the material takes place with a water saw, which works by friction with the stone, generating a powder that, when mixed with water, forms a light grey mud. All these materials, both the cutting waste and the mud have been classified as "by-products" and therefore in all respects are not waste and can be used for other production processes such as that of cement.

Figure 1–Piasentina Stone in a quarry

Figure 2–Piasentina Stone tile



A summary of the main mechanical properties of the material is reported in Table 2.

Table 2–Mechanical properties of Piasentina Stone

Volume Weight	2690	kg/m^3
Water absorption	0,21	%
Stress load (compressive)	145	MPa
Elastic tangential module	72600	MPa
Stress loas (tensile)	30	MPa

Close friction wear (reference to San Fedino granite)	0,58	-
Knoop micro-hardness	1830	MPa

EXPERIMENTAL PROGRAM

The basic idea of the research was to find a possible substitute for clinker within cement, which would comply with the limitations imposed by European legislation and at the same time would not be a virgin material, but the waste of another production chain. Observing the local rocks in the Friuli Venezia Giulia area, it emerged that the Piasentina stone contains more than 95% limestone, which makes it a material that can potentially be used; it was also considered that the waste deriving from its extraction represents a substantial part by weight, which therefore, if reused, can be considered as "recycled".

The experimentation on mixtures containing Piasentina Stone began with an analysis of the chemical composition of the material, evaluating its compatibility with the restrictions imposed by the regulations in force for limestone.

Following the European code UNI EN 197-1 [8], a material can be recognized as limestone if it has the following properties:

- The calcium carbonate ($CaCO_3$) content must be equal to or greater than 75% by mass;
- The clay content (measured by the methylene blue test in accordance with EN933-9 [11]) must not exceed 1.2g/100g (0,012 lb/lb);
- The total carbon content (TOC) must satisfy one of the following criteria
  1. LL: must not exceed 0.2% by mass;
  2. L: must not exceed 0.5% by mass.

In particular, a percentage of $CaCO_3$ equal to 95.27% was determined, exceeding the limit imposed by the regulations, set at a minimum of 75%. The TOC was measured with a value lower than 0.01% by mass, significantly lower than the limit of 0.2%, while the methylene blue test did not detect fine particles.

Figure 3–Piasentina Stone

The method for determining the material properties was the use of XRD analysis (results reported in Figure 4), of which an extract of the results is reported:

Quartz (%): 2,2%

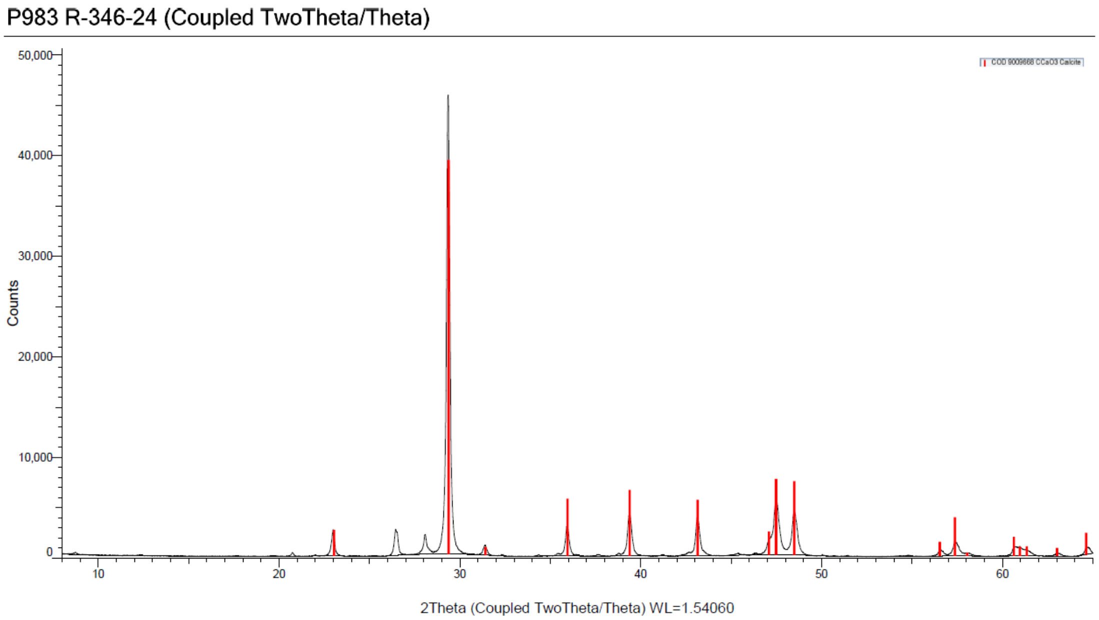
Illite (%): 1,7%

Albite (%): 0,3%

K-Feldspar (%): 0,1%

Kaolinite (%): 0,5%

Figure 4–Results of XRD analysis



Based on the data obtained, it was possible to proceed with the second phase of the research, being able to consider the Piasentina stone as "limestone" within the reference standard UNI EN 197-1.

The use of limestone is allowed both for binary and ternary cements, following the codes UNI EN 197-1, UNI EN 197-5 [9] and UNI EN 197-6 [10]. The limits are defined in Table 3, Table 4 and Table 5.

Table 3–Cements composition following UNI EN 197-1

Main types	Notation		Clinker	Blast-furnace slag	Silica fume	Pozzolana		Fly ash		Burnt shale	Limestone		Minor additional constituents
			K	S	D	natural P	natural calcined Q	siliceous V	calcareous W	T	L	LL	
CEM I	Portland cement	CEM I	95-100	-	-	-	-	-	-	-	-	-	0-5
CEM II	Portland-slag cement	CEM II/A-S	80-94	6-20	-	-	-	-	-	-	-	-	0-5
		CEM II/B-S	65-79	21-35	-	-	-	-	-	-	-	-	0-5
	Portland-silica fume cement	CEM II/A-D	90-94	-	6-10	-	-	-	-	-	-	-	0-5
	Portland-pozzolana cement	CEM II/A-P	80-94	-	-	6-20	-	-	-	-	-	-	0-5
		CEM II/B-P	65-79	-	-	21-35	-	-	-	-	-	-	0-5
		CEM II/A-Q	80-94	-	-	-	6-20	-	-	-	-	-	0-5
		CEM II/B-Q	65-79	-	-	-	21-35	-	-	-	-	-	0-5
	Portland-fly ash cement	CEM II/A-V	80-94	-	-	-	-	6-20	-	-	-	-	0-5
		CEM II/B-V	65-79	-	-	-	-	21-35	-	-	-	-	0-5
		CEM II/A-W	80-94	-	-	-	-	-	6-20	-	-	-	0-5
		CEM II/B-W	65-79	-	-	-	-	-	21-35	-	-	-	0-5
	Portland-burnt shale cement	CEM II/A-T	80-94	-	-	-	-	-	-	6-20	-	-	0-5
		CEM II/B-T	65-79	-	-	-	-	-	-	21-35	-	-	0-5
	Portland-limestone cement	CEM II/A-L	80-94	-	-	-	-	-	-	-	6-20	-	0-5
		CEM II/B-L	65-79	-	-	-	-	-	-	-	21-35	-	0-5
		CEM II/A-LL	80-94	-	-	-	-	-	-	-	-	6-20	0-5
		CEM II/B-LL	65-79	-	-	-	-	-	-	-	-	21-35	0-5
	Portland-composite cement[c)]	CEM II/A-M	80-88	12-20									0-5
		CEM II/B-M	65-79	21-35									

Table 4–Cements composition following UNI EN 197-5

Main types	Notation		Composition (percentage by mass)										
			Main constituents										Minor additional constituents
			Clinker	Blast-furnace slag	Silica fume	Pozzolana		Fly ash		Burnt shale	Limestone		
						natural	natural calcined	siliceous	calcareous				
			K	S	D[b)]	P	Q	V	W	T	L[c)]	LL[c)]	
CEM II	Composite Portland cement	CEM II/C-M	50-64	36-50									0-5
CEM IV	Composite cement	CEM VI (S-P)	35-49	31-59	-	6-20	-	-	-	-	-	-	0-5
		CEM VI (S-V)	35-49	31-59	-	-	-	6-20	-	-	-	-	0-5
		CEM VI (S-L)	35-49	31-59	-	-	-	-	-	-	6-20	-	0-5
		CEM VI (S-LL)	35-49	31-59	-	-	-	-	-	-	-	6-20	0-5

Table 5–Cements composition following UNI EN 197-6

Main types	Notation		Composition (percentage by mass)											
			Main constituents											Minor additional constituents
			Clinker	Fine fraction of recycled concrete	Blast-furnace slag	Silica fume	Pozzolana		Fly ash		Burnt shale	Limestone		
							natural	natural calcined	siliceous	calcareous				
			K	F	S	D[b]	P	Q	V	W	T	L[c]	LL[c]	
CEM II	Portland cement with recycled concrete	CEM II/A-F	80-94	6-20	-	-	-	-	-	-	-	-	-	0-5
		CEM II/B-F	65-79	21-35	-	-	-	-	-	-	-	-	-	0-5
	Composite Portland cement	CEM II/A-M	80-88	6-14	6-14									0-5
		CEM II/B-M	65-79	6-29	6-29									0-5
		CEM II/C-M	50-64	6-20	16-44									0-5
CEM VI	Composite cement	CEM VI	35-49	6-20	31-59	-	-	-	-	-	-	-	-	0-5

Preparation of the samples and definition of the test campaign

In order to evaluate the performance of the material as a substitute for clinker within the cement, therefore as a substitute for ordinary limestone in binary cements, mortar samples were made with different percentages of Piasentina Stone, to be tested at different maturation periods. In particular, a distinction is made between three mixtures: one made with only the presence of clinker, two with the presence of ordinary limestone in percentages of 5 and 10% and finally two with the Piasentina Stone in percentages of 5 and 10% (as shown in Table 6). The mixture that constitutes the samples is made up of standardized sand EN ISO679 and EN 196-1 3 parts by weight, binder 1 part by weight, water 0.5 parts by weight. Mixing and curing were carried out in accordance with the EN 196-1 [12] standard. In order to obtain comparable results and not to introduce additional parameters that could have excessively influenced the results, a constant w/c ratio of 0.5 was chosen.

Table 6–Composition of the samples

	CEM I 42,5R	Limestone	Piasentina Stone
	[%]	[%]	[%]
P989	100	0	0
P990	95	5	0
P991	95	0	5
P993	90	10	0
P994	90	0	10

On the basis of the defined combinations, it was planned to consider the resistances at 1, 2, 7, 14, 28 and 56 days, therefore in order to be able to evaluate the short and long-term behaviour of the mixtures. We did not proceed beyond 56 days because, since limestone is not a reactive substance, but is substantially inert, no long-term effect deriving from its contribution can be assumed.

Expansiveness

A common problem in many innovative mixtures is that of long-term expansivity, in fact materials that potentially guarantee a strong improvement in mechanical properties often lead to strong increases in volume in the long term, resulting, if applied for structural use, in the formation of cracks and damage. In the case of Piasentina Stone, which as a limestone behaves only as an inert material, any type of expansive phenomenon is excluded a priori, however, for safety reasons, tests were also carried out for this event. The test was therefore conducted according to UNI EN 196-3 [13] with a mixture (70% type I cement and 30% Piasentina Stone, like the verification test for fly ash) and the following results of specimen expansion were obtained: the maximum measured value was 0.6 mm (0,024 in), when the maximum acceptable expansion value of the Le Chartelier calipers is equal to 10 mm (0,39 in). The expansivity verification was therefore considered satisfied.

Workability

The second parameter that was analyzed is the workability of the material on a vibrating table: it is in fact essential that the presence of the Piasentina Stone does not lead to strong reductions in workability. In this case, in fact, it would be necessary to intervene with the introduction of fluidifiers or with an increase in the quantity of water in the mixture, at the cost, in the latter case, of a reduction in resistance. Wanting to evaluate the effects of the introduction of the stone, the test was carried out with reference to the UNI EN 7044 [14] standard for the evaluation of the consistency of mortars.

Table 7–Workability tests results

	Spreading	Difference
	[cm]	[%]
P989	18,6	-
P990	18,1	2,7%
P991	18,3	1,6%

P993	18,3	1,6%
P994	18,6	0,0%

As can be seen from Table 7, the spreading values differ from each other by less than 3% and therefore it can be considered that the Piasentina Stone has no influence on the behaviour of the mixture.

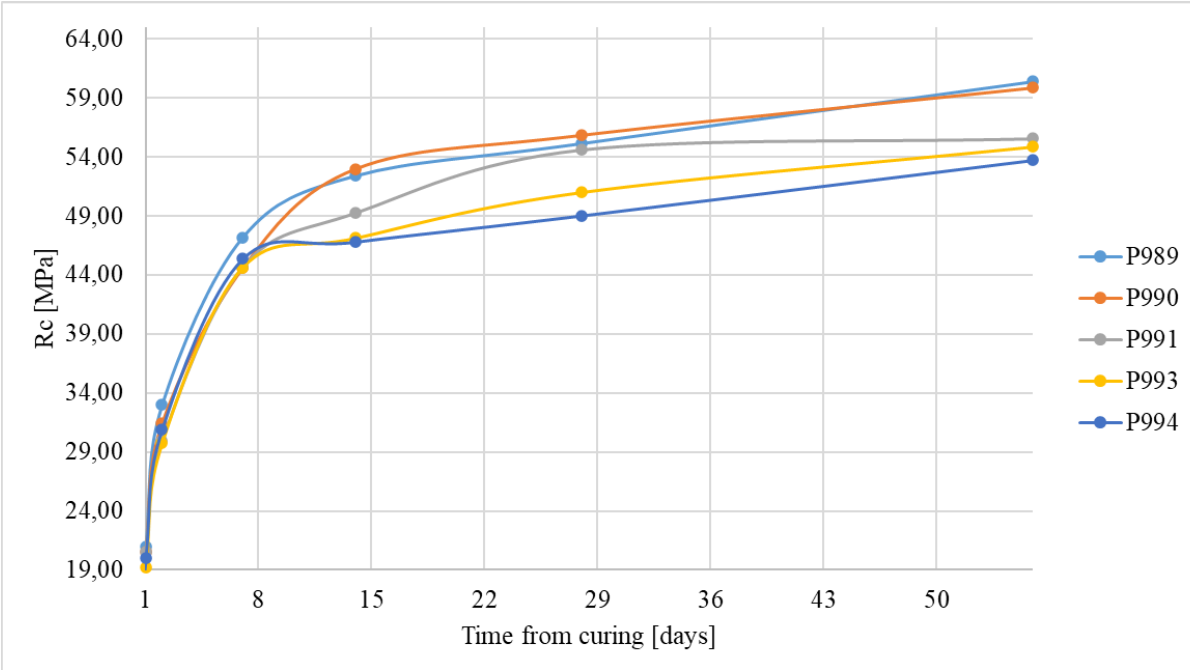
COMPRESSIVE STRENGTH TESTS

The mechanical parameter of reference for the resistance of mortars is the compression one and therefore the evaluations were conducted with respect to this value. A summary of the results obtained is reported below in table 8 and in Figure 5; for each type of mixture it was chosen to carry out the test of two specimens and taking the average resistance as the reference value.

Table 8–Compressive strength test results

Compressive strength for different time from curing [MPa]							
	Piasentina	**1**	**2**	**7**	**14**	**28**	**56**
	[%]	**[dd]**	**[dd]**	**[dd]**	**[dd]**	**[dd]**	**[dd]**
P989	0,00	20,96	32,98	47,20	52,42	55,16	60,41
P990	5,00	20,53	31,42	44,59	53,00	55,85	59,87
P991	5,00	20,52	29,92	44,70	49,28	54,62	55,57
P993	10,00	19,20	29,74	44,65	47,13	51,01	54,87
P994	10,00	20,02	30,88	45,36	46,80	49,03	53,73

Figure 5–Compressive strength test results



As can be seen from the graph, at 1, 2 and 7 days the resistances are substantially coincident, with a negligible increase in the mix without substitutions. At 14 days of maturation, a difference begins to emerge between the specimens with 5 and 10% replacement of Piasentina Stone and with 10% limestone and the reference one. At 28 days, the resistance to which it is essential to refer for the structural design, the resistance of the specimens with 5% replacement of limestone and Piasentina Stone appear to have similar performances to those of the reference mix, while in the mixes with 10% replacement a loss of approximately 10% is observed. Since Piasentina Stone and limestone are inert substances, it is not expected that in the long term there will be substantial variations in resistance, other than those due to the clinker, thus leading to a settling of the performances to an asymptotic value. This behaviour is also confirmed by the tests carried out, with a maintenance of the trend present at 28 days, with a slightly more accentuated increase in resistance for the reference mixture and the one with 5% limestone, while the other mixtures, less performing, present lower values of approximately 20%.

EXPERIMENTAL RESULTS DISCUSSION

The test campaign conducted on mortars has highlighted excellent behavior for all the mixtures, with differences that, for the reference resistance of 28 days, do not exceed 10%. In the long term, losses are more accentuated, but this parameter is not substantial for the design, as it does not lead to the variation of the parameters to be used for the calculation. It must also be considered that, if this problem is to be corrected, it is possible to resort to further additions of reagent components coming as waste from other production chains (such as rice husk ash), which guarantee an increase in long-term resistance thanks to the presence of SiO_2. In light of the results obtained, it is therefore possible to consider the use of Piasentina Stone valid in the production of cement mixtures.

COMPARISON WITH OTHER EXPERIMENTAL TESTS

In order to compare the results obtained for the Piasentina Stone with a similar case, a study conducted on mortars made with partial replacement of the clinker with basalt powder was taken into consideration [15]. In this case the composition of the material is not totally inert, since more than 50% silicon is present: this fact will be taken into consideration when comparing the results. However, since silica develops its effect in the long term (over 28 days), substantial differences are not expected compared to the case study. In the research, six different cement mixtures were created using CEM I (95% clinker and 5% gypsum), in which the clinker was gradually reduced, according to different percentages, with basalt powder. A summary table of the compositions of the samples is reported in Table 9:

Table 9–Samples composition

	Clinker	Basalt	Gypsum
Mixture	**[%]**	**[%]**	**[%]**
S0	95,5	0	4,5
S1	85,5	10	4,5
S2	80,5	15	4,5
S3	75,5	20	4,5
S4	70,5	25	4,5
S5	65,5	30	4,5

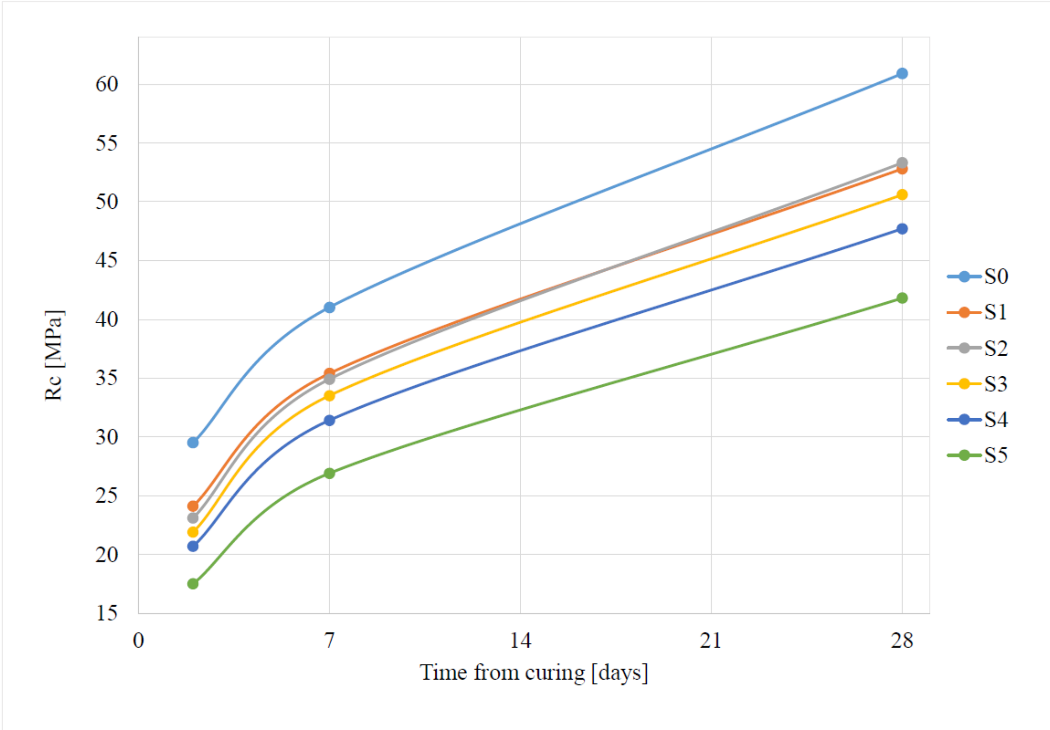
In particular, based on the analyses carried out, basalt has the following chemical composition:
SiO_2: 50,2%;
Al_2O_3: 14,2%;
Fe_2O_3: 14,4%;
CaO: 10,7%;
MgO: 7,5%;
Na_2O: 1,8%.

In the study considered, compressive strength tests were carried out on the mortars defined above at different maturation times of the mixtures: 2, 7 and 28 days, the results of which are reported in Table 10 and Figure 6.

Table 10–Compressive strength test results

Compressive strength [MPa]				
Mixture	**[%]**	**2**	**7**	**28**
S0	0	29,5	41	60,9
S1	10	24,1	35,4	52,8
S2	15	23,1	34,9	53,3
S3	20	21,9	33,5	50,6
S4	25	20,7	31,4	47,7
S5	30	17,5	26,9	41,8

Figure 6–Compressive strength test results



It is observed that the control mixture reaches the same resistance at 28 days as that considered in the experimental tests of this study, equal to about 60 MPa (12500 psf); similarly, it is evident that as the percentage of substitution increases, a reduction in resistance is evident. In the case of the Piasentina stone, the average resistance is around 54 MPa (11250 psf), which is comparable to that of the specimens with a percentage of substitution of 10 and 15% with basalt. In light of what has been obtained, the results of this second experimental campaign can be considered comforting, which can therefore "validate" the results of this study. The only difference that can be perceived is in the trend of the growth rate of the resistance curves, which in the case of basalt are more sloped between 7 and 28 days: the difference is attributable to the reactive behavior of the silica contained in the basalt, which leads to an increase in resistance starting from 7 days.

FURTHER RESEARCH

In this study, chemical analyses of the material and tests with mortars were carried out: it is essential that preventive research for the use of Piasentina Stone continues through the analysis of the behaviour of concretes. Only after carrying out these tests with different percentages of replacement of clinker with Piasentina Stone, if the results continue to prove promising, will it be possible to confirm the possibility of using this material for structural purposes.

CONCLUSIONS

Clinker production is extremely polluting and it is essential to both develop energy saving methods within cement kilns and reduce the quantity of this component within cement mixes. In light of the tests carried out, Piasentina Stone appears to be an excellent substitute (in a percentage between 5 and 10%), while at the same time ensuring a reduction in CO_2 emissions and the use of a material that would otherwise be waste. If the future campaign of tests on concrete confirms the results obtained for mortars, it will be possible to obtain highly sustainable cements that comply with the environmental requirements of recent regulations. An approach of this type, which aims to enhance the materials directly available in the territory, can be extended to all areas of Italy, leading the construction sector towards a more sustainable and circular economy.

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BIOGRAPHY

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