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Mechanical characterization of cement mortars and concrete with recycled aggregates from Coal Mining Wastes Geomaterials (CMWGs)

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A R T I C L E I N F O

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A B S T R A C T

This paper presents the results of an extensive experimental campaign related to the use of Coal Mining Waste Geomaterials (CMWGs) as recycled constituents (fine and coarse aggregates) in Ordinary Portland Cement mortars and concretes. To this purpose, a reference mix and other mixes with different percentages of replacement of natural aggregates by CMWGs, up to 40% by volume, were investigated. CMWGs came from different providers: Central Mining Institute (GIG), POLTEGOR, both in Poland, and SUBTERRA in Spain and tests were performed at two different laboratories working on similar, but not identical, compositions. This represents a novelty in the literature, generally focusing on one single-source waste and single-lab results. The physical and mechanical properties of all the mixes were evaluated and correlated with respect to the percentage of replacement of natural aggregates by CMWGs. While the presence of CMWGs, likely because of their grain size distribution, reduced the porosity of mortars (decrease of 9.5 and 20.4% for 10 and 20% of replacement respectively) and concretes (70% reduction for concretes with 10% of fines and 30% of coarse aggregates replaced by CMWGs), the mechanical properties decreased when natural aggregates were replaced with CMWGs, likely because of the reduced strength of the CMWGs aggregates. This decrease was found to be roughly proportional to the percentage of replacement of aggregates (for instance, a 12–23% reduction of flexural strength in mortars with 25% replacement of sand and, a decrease of 25% in concretes for a 25% replacement of fine and coarse aggregates); nonetheless the concrete performance remained in the range of applicability for several civil engineering applications without affecting their functionality. In conclusion, the replacement of natural aggregates by CMWGs has resulted an interesting option for real applications providing an added value to the implementation of circular economy concepts in the management and up-cycling of coal mine tailings and CMWGs.

1. Introduction

Throughout the past decades, the construction industry has been facing several challenges, including depletion of natural resources, increased energy consumption and carbon emissions, and production of excessive waste materials which directly affect the environmental equilibrium and of which it has been deemed as one of the major responsible. This has motivated the whole sector towards a deep rethinking of its traditional material production processes [1]. Fostering the uptake of circular economy procedures in the production and use of construction materials, as well as in the management of buildings and structures, mainly in the dismissal stage at the end of their service life, can lead to a paradigm change [2,3] and can contribute to reducing the environmental burden of the construction sector. Moreover, the depletion of raw material resources can be limited and the consequent consumption of soil, also in the use of landfill for the waste management. As a matter of fact this last aspect has a dramatic social importance since it can counteract with the use of soil not only for construction of dwellings but also for agricultural use and hence for the production of food.

Coal mining wastes currently stockpiled mostly in open air can represent a potential source of raw materials for the production of concrete and cementitious composites for a broad variety of uses [1].

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This attention has grown steadily since several European countries, such as Poland, have to reckon with the problem of already disposing up to 812 Mtons of Coal Mine Waste Geomaterials (CMWGs) in disposal sites and/or storage facilities, being the annual volume of mining extractive waste currently generated in Poland up to 30 Mtons [4].

The solid Coal Combustion Waste (CCW), also known as coal ash, contains large quantities of contaminant metals and is one of the largest solid wastes produced in the world. There are several different methods of reutilization of CCW, including cement production, backfill for open voids, landscaping materials and revegetation of substrate at mine sites [5]. This research focuses on the reutilization of solid Coal Mine Waste Geomaterials (CMWGs) in concrete production, both as replacement of cement and of natural aggregates. A particular application was investigated by Zhang et al. [6] finding that foam concrete mixed with coal gangue can exhibit good mechanical properties with a reduced porosity; thereby, coal gangue foam concrete can be used for the confinement filling or working faces in mined-out areas.

Modarres et al. [7] analyzed cementitious composites produced by substituting cement with the three different types of coal waste recycled materials: coal waste powder (CWP), coal waste ash (CWA) and CWA together with limestone powder (CWA-LS). The compressive strength results showed that the concrete mixtures containing coal materials at a 5% replacement level (by mass) had higher compressive strength compared to the Ordinary Portland Cement (OPC) concrete, likely because of the filler effect of the fine coal waste powder particles. Moreover, an increased toughness of the blended-cement concretes compared to the reference one was observed when cement was replaced with coal waste powder and coal waste ash up to 10% (always by mass). This could be attributed, as before, to the filler effect of the coal waste powder particles.

Vegas et al. [8] also studied the effect of replacing cement with CMWGs at different percentages, namely 0%, 6%, 10%, 20% (by mass). They found that the addition of mining waste slightly accelerates setting times and results into a loss of workability, likely because of increased fineness of CMWGs. Moreover, compressive and flexural strengths were evaluated at different ages, up to one year. The authors argued that metakaolin contained in activated CMWGs resulted in early pozzolanic reaction, which fostered earlier strength development but whose effect tended to be smoothed over time; a similar trend was hypothesized for the reactive carbonated phases in the CMWGs, that could have promoted an earlier stronger adhesion between the aggregates and the paste.

Drying shrinkage increased with the level of replacement of OPC by CMWGs.

Frías et al. [9] studied the effect of thermally ACMW at 650 °C on the properties of blended cements: after calcination under controlled conditions, the coal waste showed high pozzolan activity due to the transformation of the kaolinite contained in the CMWGs into metakaolin, allowing it to be used in the production of blended cements (commercially known as type II/A cements).

Caneda-Martinez et al. [10] studied how the presence of thermally ACMW in concrete affected steel corrosion related to chloride ion content. A chloride-induced accelerated corrosion test was conducted in steel bars embedded in four different mortar specimens: a reference one and three others with partial substitution of ordinary Portland cement by activated coal mining waste (substitutions of 10%, 20% and 50% by mass). The addition of ACMW to concrete induced a decrease of the critical chloride ion content up to 90% when compared to the reference specimens. Mixes with coal mining waste had a longer corrosion onset time, due to higher resistance to chloride ion penetration and lower chloride diffusion coefficients due to microstructure densification.

A different concept was employed by Okagbu et al. [11], who investigated the effectiveness of Portland cement in the stabilization of Nigerian coal-reject, to be further used in construction. Shrinkage decreased with increasing coal waste percentage probably due to the reactions between the coal waste fines and the cement leading to the formation of coarse particles. The results of unconfined compression tests showed an expectable increase in compressive strength with increasing amount of cement.

Whereas the previous studies focused on the replacement of cement with fine CMWGs, a few studies have investigated the possibilities of employing even larger fractions of less fine CMWGs for the replacement of natural sand. Wu et al. [12] prepared two series of concrete with different replacement levels (25%, 50%, 75%, and 100%) of sand and aggregate by CMWGs. They found that the compressive strength and density of concrete containing coal mine aggregates decreased with the increase of the replacement level of natural aggregates.

Santos et al. [13] studied the substitution of fine aggregates with coal mining waste from Brazilian mines in concrete paving blocks. It was concluded that the production of satisfactory concrete pavement blocks is possible by substituting the natural river sand aggregates by coal mining wastes in volume replacement percentages up to 50%.

The total replacement (100%) of natural sand with coal mine aggregates was also investigated by other researchers [13–20]. Specifically, Singh et al. [19] observed that with a total replacement (100%) of fines with coal bottom ash, the dry bulk density of concrete mixtures decreased of around 10%. Muthusamy et al. [18] found that the total replacement with bottom ash can cause a decrease of 24% in flexural strength compared to a reference concrete.

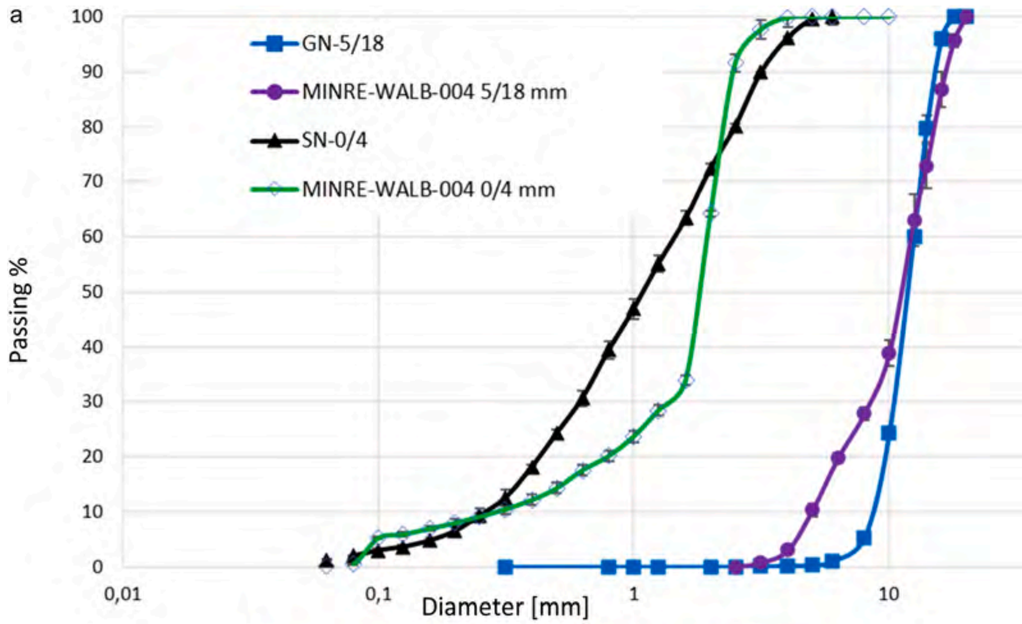
Few other studies on concrete made with coal mining waste are present in literature so far [1] which have investigated the feasibility of using CMWGs as both fine and coarse aggregates in cement-based concrete [14,17,19,21,22], confirming they can be good candidates as secondary raw materials for construction purposes [1].

A few investigations have also assessed the deterioration of the mechanical properties of concrete due to the presence of CMWGs. According to Zhang et al. [23], concrete gas permeability was significantly reduced by the addition of coal gangue.

Within this context, this paper presents a comprehensive experimental campaign to evaluate the use of CMWGs as recycled fine and coarse aggregates in cement mortars and concretes as replacement of natural aggregates in different percentages. To this purpose, mortars and concretes with CMWGs have been produced by two universities and employing as above, CMWGs from different sources: Politecnico di Milano (PoliMi) and CY Cergy Paris University (CYU) as detailed below, in the attempt of validating a circular economy valorization concept for the management of CMWGs for operating and (to-be) dismissed coal mine sites. The involvement of two different laboratories, working on complementary mix compositions, and employing materials from different sources stands, to the authors' best knowledge, as a one-of-a-kind work to validate a performance based approach to the upcycling of secondary raw materials in the production of (cement based) construction materials and products. The investigation, developed in the framework of MINRESCUE project funded by the European Commission within the Research Fund for Coal and Steel (RFCS) (GA 860006), intends to pave the way not only to implement circular economy concepts in the management of wastes of coal mining activities (also in the sight of a dismissal of fossil fuel energy production) but also to validate a comprehensive performance based methodology for a broader uptake of circular economy practices in the construction industry overcoming the drawback of the variability in the waste/secondary raw material supply chain. The variety of the tests performed at three different laboratories and the provenance of the secondary raw Coal Mine Waste Geomaterials from four different sources adds value to the study but also constitutes a novelty itself, providing a representative basis for the validation of the novel use CMWGs as aggregates in concrete production.

2. Materials selection and characterization

PoliMi and CYU investigated companion concrete mixes with the same constituent materials whereas the CMWGs used as recycled aggregates were provided by different partners of the MINRESCUE project, including the Central Mining Research Institute (GIG) from Katowice a



b

Fig. 1. (a) Particle size distribution of employed natural and CMWGs aggregates; (b) CMWGs aggregates.

(Poland), SUBTERRA Engineering from Madrid (Spain) and POLTEGOR Institute of Opencast Mining from Walbrzych (Poland).

The constituent materials for concrete were the same for both universities and they were the following:

- Cement II/A-L 42.5 R with a specific gravity of 3.05 provided by Superbeton, Ponte della Priula, Italy.
- Limestone filler DOP N1040 with a specific gravity of 2.72 provided by Bernardelli, Italy.
- Carbonate and quartz-rich sand SN 0/4 with particle size ranges from 0 to 4 mm.
- Carbonate and quartz- natural rounded gravel GN 5/18 with particle size ranges from 4 to 18 mm.
- Superplasticizer DYNAMON NRG1030 ®, specific gravity 1.04.
- On the other hand, each university used CMWGs from different providers:
- CYU used fine and coarse aggregates from CMWGs provided by POLTEGOR: CMWGs sand, named MINRE -WALB-004 0/4 mm and CMWGs gravel, named MINRE -WALB-004 5/18 mm, obtained by mixing 25% of G-WC- 4/10 and 75% of G-WC-10/18.
- Regarding PoliMi, for mortars, fine CMWGs were provided by the Central Mining Institute (GIG). For concretes, fine CMWGs were provided by GIG and SUBTERRA, whereas the coarse CMWGs aggregates were provided by GIG, SUBTERRA and POLTEGOR. The CMWGs provided by POLTEGOR came from Walbrzych (Poland). Those provided by GIG came from also from Poland, specifically from Radlin, Bierun and Rybnik, whereas those provided by SUBTERRA came from Ponferrada (Spain).

An example of grain size distribution of both natural and CMWGs aggregates is shown in Fig. 1a, with reference to CMWG sand labelled MINRE -WALB-004 0/4 mm and CMWG gravel, named MINRE -WALB-004 5/18 mm, obtained by mixing 25% of G-WC- 4/10 and 75% of G-WC-10/18. It can be observed that the natural sand contains more fines than CMWGs and the opposite is happening for the coarse aggregate fraction. This information was duly considered in the optimization of the granular solid skeleton of the concrete mixes containing CMWGs. Similar considerations hold for CMWGs coming from other sources. Fig. 1b shows the CMWGs used in the study.

Moreover, according to Hazen's uniformity coefficient (C_U) and curvature coefficient (C_C), the sand can be considered as well graded

Table 1
Physical properties of the aggregates.

Property	Standard	SN 0/4	MINRE -WALB-004 0/4 mm	GN-5/18	MINRE -WALB-004 5/18 mm
Bulk density (kg/m^3)	EN 1097–6	2.68	2.17	2.7	2.16
$WA_{24\ h}$	EN 1097–6	1.4	2.6	1.1	7.2
Fineness modulus	EN 933–3	3.5	3.8	-	-
C_U	EN 933–3	6.4	6.3	1.6	1.5
C_C	EN 933–3	1	2.4	1	1

Table 2
Characterization test results of the CMWGs used in this research.

Parameter	Content as received	Dry content
Water (%m/m)	1.03	-
Ash (%m/m)	84.83	85.71
Carbon (%m/m)	5.89	5.95
Hydrogen (%m/m)	0.22	0.23
Sulphur (%m/m)	0.35	0.35
Organic carbon (%m/m)	5.89	5.95
Gross calorific value (J/g(kJ/ka)	1400	1410
Net calorific value (J/g(kJ/ka)	1320	1360
Specific density (g/cm^3)	2.64	-
Particle size distribution		
D10 = 5.0 mm D50 = 8.5 mm		**D90 = 10.2 mm**

Table 3
Mortar and concrete mixes produced in this research.

Type	University	Mix ID	CMWG provider	% replacement of natural aggregates by CMWG
Mortar	PoliMi	Reference mortar	-	-
		Mortar-25%	GIG	25% (sand)
Mortar	CYU	Reference mortar	-	-
		Concrete Equivalent Mortar-10%	POLTEGOR	10% (sand)
		Concrete Equivalent Mortar-20%	POLTEGOR	20% (sand)
Concrete	PoliMi	Reference	-	-
		FO-GIG	GIG	25% (sand)
		FO-SUB	SUBTERRA	25% (sand)
		FO-GIG+SUB	GIG/ SUBTERRA	25% (sand)
		CO-GIG	GIG	25% (gravel)
		CO-SUB	SUBTERRA	25% (gravel)
		CO-POL	POLTEGOR	25% (gravel)
		FC-GIG	GIG	13% (sand) + 13% (gravel)
		FC-SUB	SUBTERRA	13% (sand) + 13% (gravel)
		FC-POL	POLTEGOR	13% (sand) + 13% (gravel)
Concrete	CYU	Reference	-	-
		C-F10%-G30%	POLTEGOR	10% (sand) +30% (gravel)

(C_U>6 and C_C=1), whereas all the gravel appears to be poorly graded ($C_U \cong$ 1). The properties of aggregates, water absorption at 24 hours ($WA_{24\ h}$) and bulk density obtained according to the EN 1097–6, are summarized in Table 1, highlighting a lower specific gravity and a much higher water absorption of CMWGs as compared to natural aggregates, which will obviously reflect in the properties of the investigated concretes incorporating them as aggregates.

Table 4
Reference concrete mix composition.

Material [kg/m^3]	PoliMi	CYU
Cement II 42.5	330	326
Sand 0/4	950	938
Gravel 5/18	890	879
Filler DOP N1040	60	59
Superplasticizer Dynamon NRG 1022	2	2
Water (w/c)	160 (0.48)	158 (0.48)

Table 5
Mix composition of concretes using CMWGs developed by CYU and PoliMi.

Constituent (kg/m^3)	Fine and coarse mix CYU	Fine only mix (FO) PoliMi	Coarse only mix (CO) PoliMi	Fine and coarse mix (FC) PoliMi
Cement CEM II 42.5	326	330	330	330
Natural sand 0/4	844	723	964	839
Filler DOP N1040	59	77	77	77
Natural gravel 5/18	615	896	672	780
SP Dynamon NRG 1022	2	1.98	1.98	1.98
CMWG fines 0/4	78	241	-	125
CMWG coarse 5/18	200	-	224	116
Water	158	158	158	158

Mine waste samples were tested in the GIG laboratory. Table 2 summarizes the results of the following tests performed: specific gravity, initial water content, optimum water content, dry and bulk densities, petrographic composition, ash content, density analysis and sulfur content, caloric, moisture, coal content. Moreover, Table 2 includes the grain characteristics of the waste based on the determination of the diameter: diameter corresponding to 10%, 50% and 90% passing (D10, D50, D90 respectively).

3. Experimental programme: materials and methodologies

Table 3 summarizes the experimental plan developed in this study. Both universities CYU and PoliMi cast the same reference concrete and different concrete and mortar mixes with different levels of replacement of natural aggregates by CMWGs. As shown in Table 3, CYU made a concrete with a replacement of 10% for natural sand and 30% for natural gravel by CMWG aggregates from POLTEGOR, whereas PoliMi developed three different concrete mixes replacing 25% by volume of natural aggregates by CMWGs from different origins. Specifically, the following concrete mixes were produced: a fine only mix (FO) replacing a 25% of natural sand by fine CMWGs provided by both GIG and SUBTERRA (one mix with each aggregate), as also defined in Table 3; a coarse only mix (CO) in which a 25% of volume of raw gravel was replaced by coarse CMWGs from GIG, SUBTERRA and POLTEGOR; and a fine a coarse mix (FC) with a substitution of both sand and gravel by fine and coarse CMWGs, respectively at 13% by volume for each fraction also provided by GIG, SUBTERRA and POLTEGOR.

The mix design of the reference concrete was based on the one provided by Nuova Tesi, an Italian company of precast concrete elements, partner of the MINRESCUE project. The mix design is shown in Table 4 and is currently employed in the production of precast cladding panels. CYU considered the air content in the fresh concrete, equal to 0.02 m^3, which resulted into a slightly different reference mix design. The concrete mix designs using CMWG produced at CYU and POLIMI are shown in Table 5.

In addition to concretes, both universities also investigated mortars:

Table 6
Mix composition of mortars using CMWGs developed by CYU.

Constituent (kg/m^3)	Reference mortar	Mortar-10%	Mortar-20%
Cement II 42.5	486	486	486
Natural sand 0/4	1463	1314	1168
Waste sand 0/4	0.0	118	236
Filler DOP N1040	88	88	88
SP DYNAMON NRG1030	2.0	2.0	2.0
Water	236	236	236

Table 7
Mix composition of mortars using CMWGs developed by PoliMi.

Constituent (kg/m^3)	Reference mortar	Mortar 25%
Cement 42.5	510	510
Natural sand 0/4	1489	1117
Filler DOP N1040	119	119
SP Dynamon NRG 1022	3	3
CMWG sand	-	372
Water	244	244

PoliMi cast a mortar with a 25% replacement of natural sand by fine CMWGs from GIG whereas CYU casted two mortars replacing a 10% and 20% of natural sand by fine CMWGs provided by POLTEGOR. Table 6 and Table 7 summarize the mix compositions of all the investigated mortars for CYU and PoliMi respectively. Mortar mix designs were obtained eliminating the coarse aggregates from the original concrete mix and scaling up the constituent proportions to a unit volume.

The adopted casting procedure, based on the experience at PoliMi, consisted of the following steps:

- the gravel (and coarse CMWG, if any), sand (and fine CMWG, if any) and cement were added in the mixer, in this specific order and were dry-mixed for 2 minutes;
- the limestone filler was then added and the mixer was covered with a plastic sheet and turned on for around 1 minute;
- all the water was added and everything was mixed for 3 further minutes. The amount of added water was the same for all mixes (reference and with CMWGs) and, although the water absorption was different for each type of CMWGs, the water was not corrected due to the high variation of sources of supply of the different CMWGs and the unknown variability of their water absorption even inside the same batch.
- finally, the superplasticizer was added and the mixer was turned on for another 5 minutes.

The ready mixture was then transferred into oiled formworks and vibrated. One day after casting specimens were demolded and kept until testing into a relative humidity (RH) 95% and 20°C moist room. Table 8 summarizes the experimental tests performed in framework of this study.

In order to better understand the mechanical behaviour of the investigated formulations, selected concrete samples were characterized concerning their mineralogy and micro-texture at the Camborne School of Mines, University of Exeter, UK. The investigated samples, produced at PoliMi, comprised the reference sample (REF), three samples using CMWMs as coarse aggregate (CO-POL, CO-GIG and CO-SUB) and one sample using CMWMs as fine and coarse aggregate replacement (FC-SUB). The mineralogy of the aggregate used in the CO-GIG sample has been described by Nash et al. (2022) [24].

Petrographic examination of concrete polished thin sections was undertaken using a Nikon Eclipse E600 POL binocular microscope (with transmitted and reflected illumination), equipped with a Nikon Digital Sight 5MP camera.

False colour mineral maps of the investigated thin sections were obtained by collecting Energy-Dispersive X-ray Spectra (EDS) from

Table 8
Experimental Tests programme.

Mix	University	Test (standard)	Specimens (geometry and dimensions)	Curing days
Mortar	CYU	Slump EN 1015–2/3	-	28 days
	CYU	Flow table EN 1015–2/3	-	28 days
	CYU	Initial and Final Setting Time EN 196–3 (2009)	-	28 days
	CYU	Porosity ISO 15901–1	-	28 days
	CYU	Bulk density EN 1097–6	-	28 days
	CYU	Elastic modulus EN 13412	Cylinders (70×140mm)	28 days
	CYU	Compressive strength EN 196–1	Cylinders (70×140mm)	3, 7 and 28 days
	CYU	Flexural strength EN 196–1	Prisms (40×40×160mm)	3, 7 and 28 days
	POLIMI	Shrinkage EN 12617–4	Prisms (40×40×160mm)	1–320 days
	POLIMI	Compressive strength EN 196–1	Halves of prisms (40×40×80mm)	1, 3, 7, 14, 28 and 60 days
	POLIMI	Flexural strength EN 196–1	Notched prisms (40×40×160mm)	1, 3, 7, 14, 28 and 60 days
Concrete	CYU	Porosity ISO 15901–1	-	28 days
	CYU	Compressive strength EN 12390–3	Cylinders (110×220mm)	7, 14 and 28 days
	CYU	Splitting tensile strength EN 12390–6	Cylinders (110×220mm)	7, 14 and 28 days
	CYU	Elastic modulus EN 12390–13	Cylinders (110×220mm)	28 days
	POLIMI	Rheometer	20 liters batch	-
	POLIMI	Calorimeter	Cubes (100×100×100mm)	0–70 hours
	POLIMI	Shrinkage EN 12617–4	Prisms (100×100×500mm)	1–350 days
	POLIMI	Elastic modulus EN 12390–13	Cylinders (100×300mm)	10 and 28 days
	POLIMI	Compressive strength EN 12390–3	Cylinders (100×300mm)	10 and 28 days
	POLIMI	Compressive strength EN 12390–3	Cubes (100×100×100mm)	1, 2, 3, 7, 28, 60 and 90 days
	POLIMI	Flexural strength and fracture energy	Notched prisms (100×100×500mm)	7 and 28 days

samples and identifying minerals from their chemistry. This was done using a QEMSCAN® 4300 (Goodall and Scales, 2007) [25]. Sample measurement and data processing used the software packages iMeasure 4.2SR1 and iDiscover 4.2SR1 and 4.3 (Rollinson et al., 2011) [26]. The QEMSCAN® 4300 used the default settings of 25 kV, 5 nA, a 1000 X-ray count rate per pixel, a WD of around 22 mm under high vacuum and beam calibration every 30 minutes. X-ray resolution/pixel spacing was 10µm at x44 magnification. Minerals were identified from their chemical spectra. Boundary phase processors were used to alleviate edge effects and remove rogue pixels. Notably the coal/organic particles in

Table 9
Slump and flow test results for CYU mortars.

Mortar	Slump diameter (mm)	Flow table diameter (mm)
Reference mortar	136 ± 4	243 ± 4.9
Mortar-10%	123 ± 2	230 ± 1.4
Mortar-20%	117 ± 2.1	215 ± 2

Table 10
Setting times for CYU mortars *(standard deviation between parentheses)*.

Setting Time	Reference mortar	Mortar-10%	Mortar-20%
Initial Setting Time (min)	72.5 (3.5)	75 (7.1)	65 (7.1)
Final Setting Time (min)	232.5 (3.5)	279 (1.4)	305 (7.1)

Table 11
Physical properties of CYU mortars (*standard deviation between parentheses*).

	Reference mortar	Mortar-10%	Mortar-20%
Porosity (%)	14.7 (1.6)	13.3 (1.3)	11.7 (1.5)
Bulk density (kg/m^3)	2182 (0.5)	2132 (1.5)	2111 (0.5)

Table 12
Elastic modulus of CYU mortars (*standard deviation between parentheses*).

	Reference mortar	Mortar-10%	Mortar-20%
E_d (GPa)	27.9 (0.12)	25.6 (0.05)	24.6 (0.15)

these samples are not detectable using QEMSCAN® as they have a similar BSE signal to the mounting resin (Rollinson, 2021) [27]. The data therefore represents everything except the organic components.

4. Results and the main findings

In this Section, the results obtained from the tests described in Table 8 are going to be shown and analyzed, in order to validate the performance based mix-design procedure described above.

4.1. Mortar properties

•Slump and flow table tests

A very slight reduction on workability was observed when CMWGs were added (Table 9) which could be explained by the higher absorption of water of CMWGs sand compared to natural sand.

•Initial and final setting time

Adding fine CMWGs to the cement mortar has a limited effect on the initial setting time, especially for lower replacement percentages, whereas increases have been detected in the final setting times as compared to the reference ones (Table 10), for the investigated percentages of replacement. The reduction in initial setting times is consistent with literature findings and rightly attributable to the earlier reactivity of fine CMWGs. The reduction in the final setting time could be explained considering that in the investigated mixes no correction in the added water was made to account for the higher absorption of the CMWGs, which could have altered prompt water availability and hence prolonged the hydration reactions and the setting.

•Porosity and bulk density

The porosity decreased as the CMWGs aggregate replacement ratio increased, which can be explained with the fact that CMWGs aggregates have a significant role not only in filling the pores of concrete but also refining the pore structure, due to their higher content in fines and extra fines (see Fig. 1). Moreover, as expectable, given the lower density of CMWGs as compared to natural aggregates, the density of the mortar decreased with the sand replacement by fine CMWGs (Table 11).

- **Dynamic modulus of elasticity (E_d)**

The elastic modulus at 28 days decreased with the increase of natural sand replacement with fine CMWGs (Table 12), likely because of the lower stiffness of the latter as compared to natural sand.

•Shrinkage

PoliMi measured the shrinkage of mortars for 320 days (almost one year) by means of prismatic specimens (40×40×160 mm). The reference specimens had a slightly higher shrinkage compared to the ones made with CMWGs (fine only, FO, mix). Anyway, the results are similar for both type of mixes because the porosity has been reduced and hence it could compensate a higher porosity effect of the aggregates. Moreover, for each type of mix the results were very homogeneous with a very low variation of shrinkage between different specimens of a same batch (Fig. 2).

•Compressive strength

Compressive strength of mortars was determined by both universities. CYU tested cylinders 70×140mm whereas PoliMi tested the halves of prismatic specimens (40×40×80 mm) obtained after testing them by flexure as previously indicated in Table 8. The values obtained from both universities cannot be directly and immediately compared

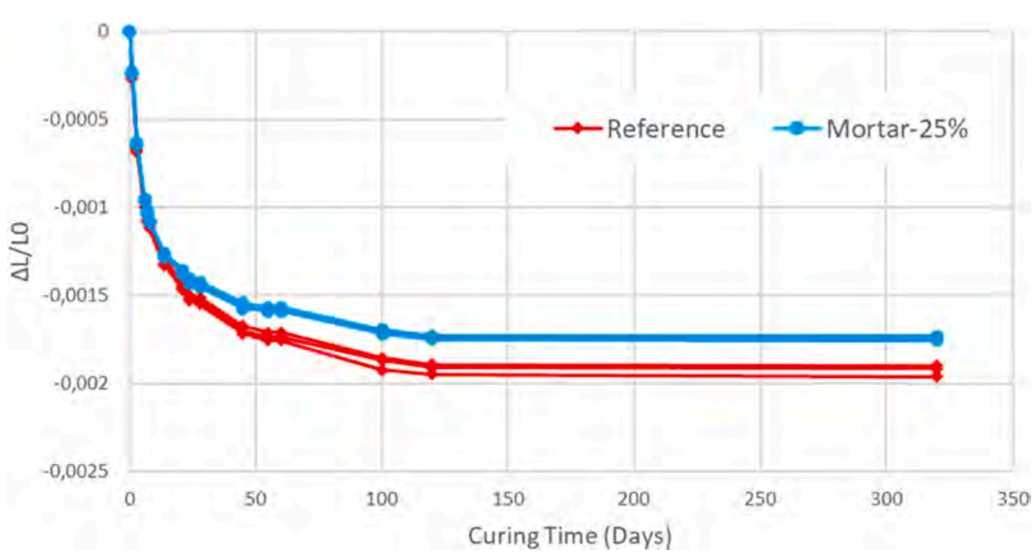


Fig. 2. Shrinkage measured on mortar specimens.

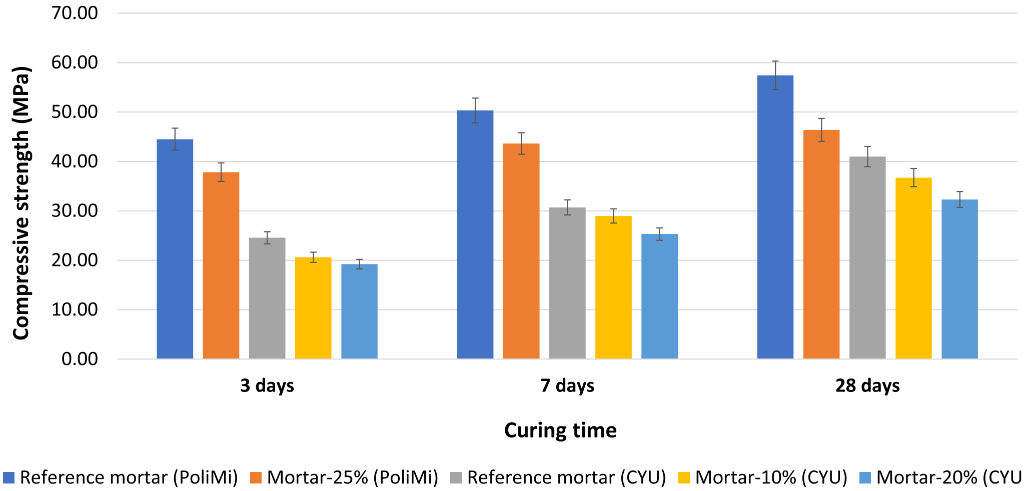


Fig. 3. Compressive strength of cement mortars versus curing time.

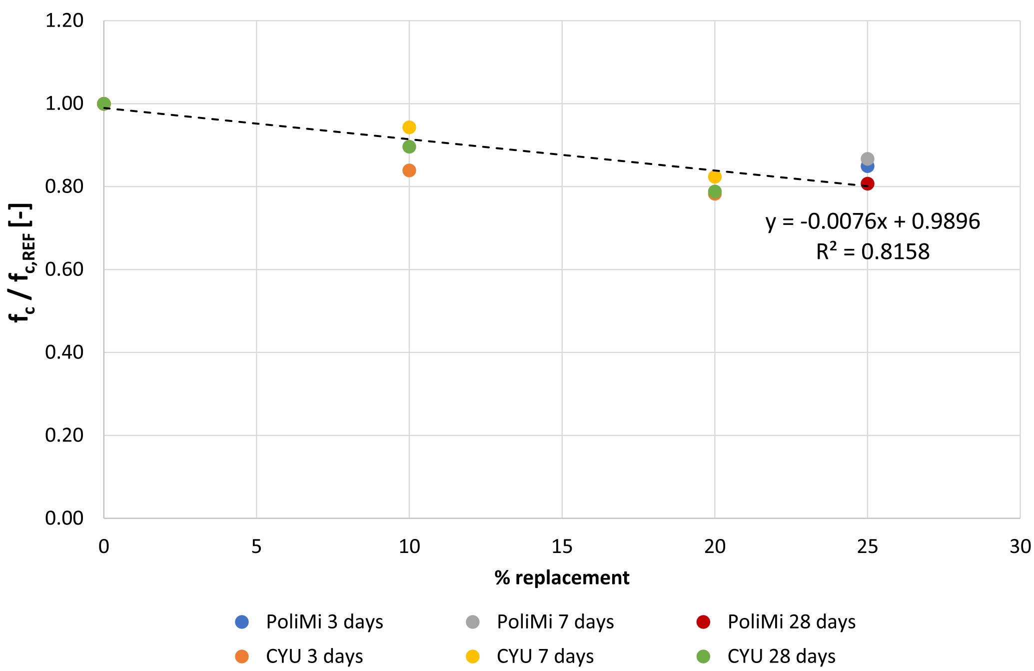


Fig. 4. Compressive strength of cement mortars versus % of replacement of natural aggregates by CMWGs.

since they were tested using different types of specimens, with different shape and scale. Anyway, for the reference samples and the same age, PoliMi obtained a higher compressive strength compared to CYU. This fact could be expected since PoliMi used prismatic specimens applying the load in a cubic surface which has a higher friction and confinement compared to the cylinders tested by CYU. PoliMi tested mortar samples after 1, 3, 7, 14, 28 and 60 days, whereas CYU tested after 3, 7 and 28 days. Fig. 3 shows the compressive strength values at the ages tested by both universities, this is 3, 7 and 28 days. For both cases, the compressive strength values decreased when fine CMWGs were added into the mix.

Considering the previously mentioned differences in the testing specimen geometries, the compressive strength values have been normalized by the compressive strength value of the corresponding reference mortar. In this way, the compressive strength values from both universities can be compared. As a matter of fact, Fig. 4 shows the dimensionless compressive strength ($f_c/f_{c,REF}$) of the tested mortars versus the percentage of replacement of natural aggregates by CMWGs. The results of all the tests performed at either laboratory are well correlated as shown by the linear trend in Fig. 4 with a quite good coefficient of correlation ($R^2 > 0.8$). In any case, the decreasing trend is less than proportional with respect to the percentage of replacement.

4.2. Flexural strength

Both universities conducted flexural tests on prismatic specimens 40×40×160mm. Fig. 5 shows the flexural strength values at 3, 7 and 28 days. The mortar with 25% of CMWGs cast at PoliMi featured a lower flexural strength (12–23%) compared to the reference mortar. On the other hand, CYU mortars with 10 and 20% of fine CMWGs were less influenced by the presence of CMWGs, especially after 3 and 7 days of curing, as shown in Fig. 5, with reductions comparable to the standard variation of flexural strength values from different specimens of the same batch. These results are also confirmed by Fig. 6 which represents the dimensionless flexural strength, calculated in a similar way to what done for compressive strength.

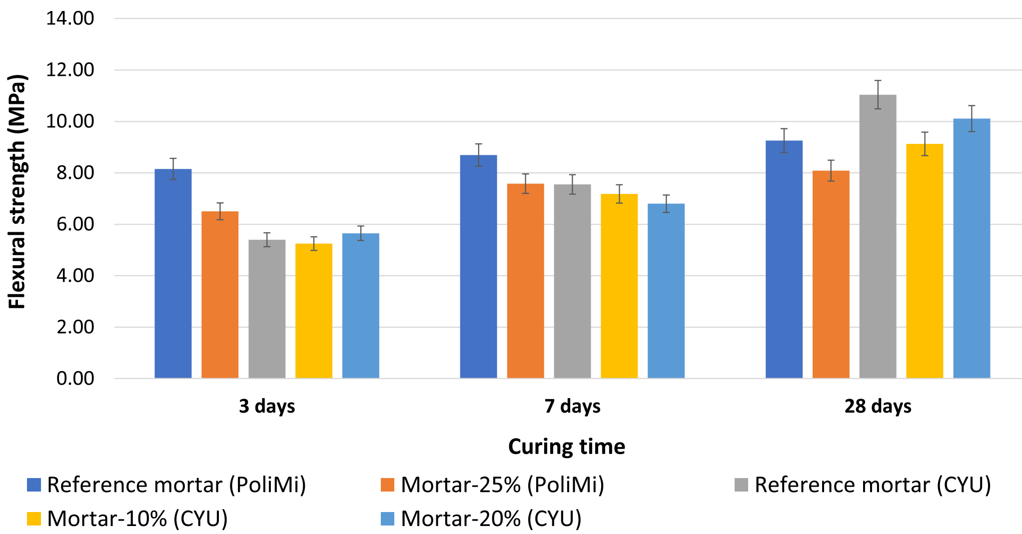


Fig. 5. Flexural strength of cement mortars versus curing time.

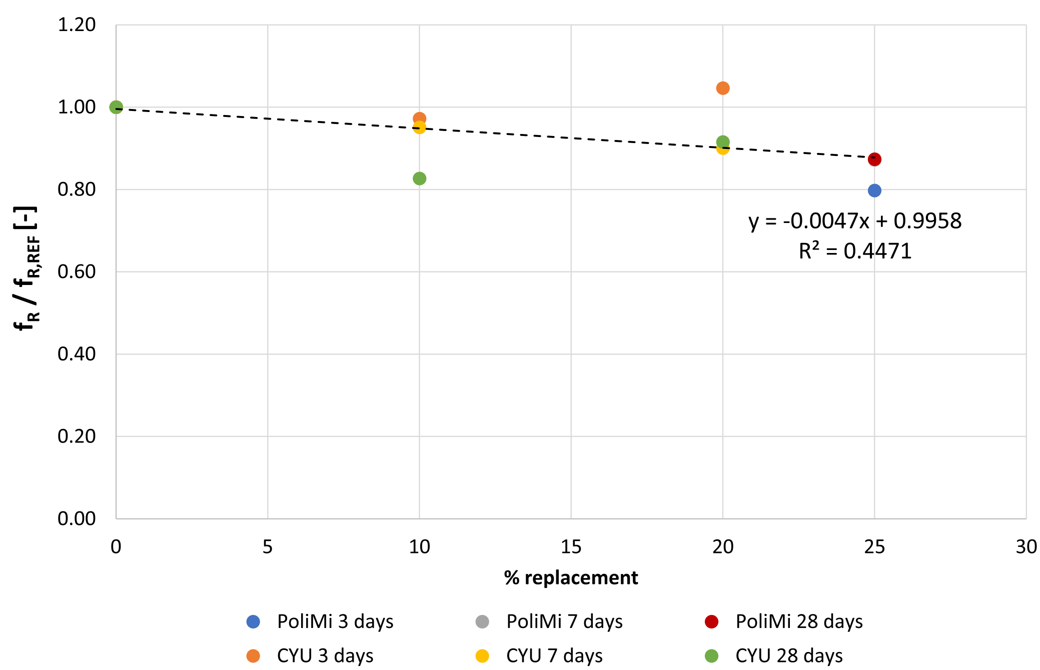


Fig. 6. Dimensionless flexural strength of cement mortars versus % of replacement.

The "decay rate" of the flexural strength with respect to that in compressive strength is even lower (slope of −0.005 approximately compared to −0.008) for fine aggregates only. This could be explained hypothesizing that more porous aggregates, like CMWGs, could act as crack arrestors and there could also be a little toughening of the aggregates paste interface even for fine aggregates due to the higher percentages of fines contained in the CMWGs.

4.3. Concrete properties

•**Rheological behavior**

The fresh state performance of concretes investigated at PoliMi was evaluated by means of rheometer tests, which allowed to identify the fundamental Bingham rheological parameters, namely plastic viscosity and yield stress. The measurements evaluated the flow behavior with an ICAR PLUS CONCRETE RHEOMETER from Germann Instruments. The first step was to fill the container of the rheometer with concrete: to ensure a proper compaction, the container was filled in two stages, tamping each layer with the steel rod used for the slump test.

Then, the rheometer tests were performed according to both the Stress Growth and the Flow Curve protocols. Both tests allow evaluating the yield stress, but only the flow curve test allows determining the plastic viscosity. Moreover, the stress growth test indicates the flowability of the mix at the beginning of the mixing process, whereas the flow curve test is used to measure the relationship between shear stress and shear rate ant to determine the Bingham parameters of yield stress and plastic viscosity. The yield stress measured with the flow curve test is the dynamic yield stress because it is measured after the breakdown of the effects of thixotropy.

Fig. 7 shows that, for a same speed, the torque applied for FO-GIG+SUB and FC-POL mixes was higher than for the reference concrete. Additionally, as shown in Fig. 8a-b (Stress Growth test and Flow curve test respectively), the yield stress and plastic viscosity were higher than the reference mix only for the following mixes: FO-SUB and FO-

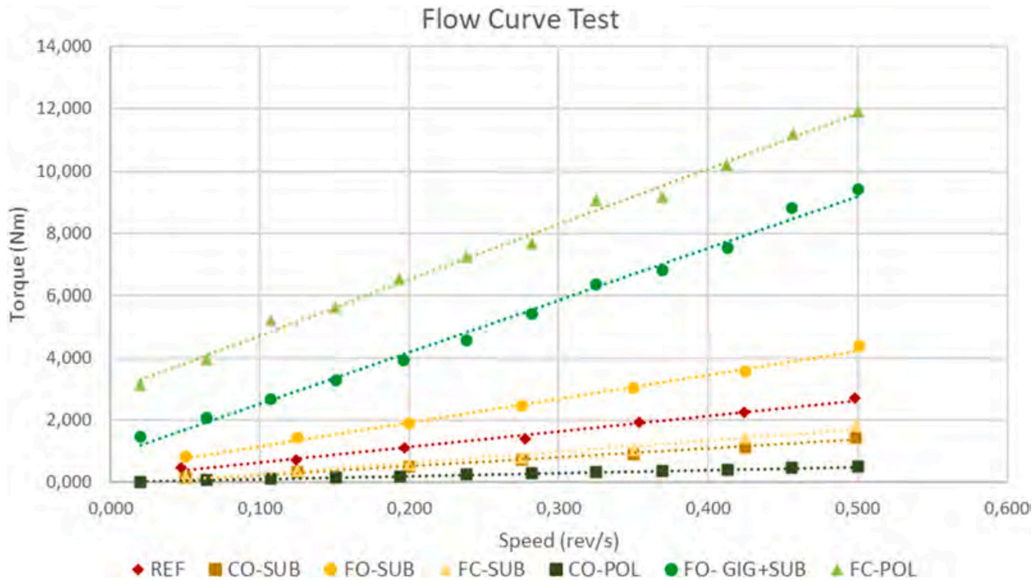


Fig. 7. Torque (N·m) versus Speed (rev/s).

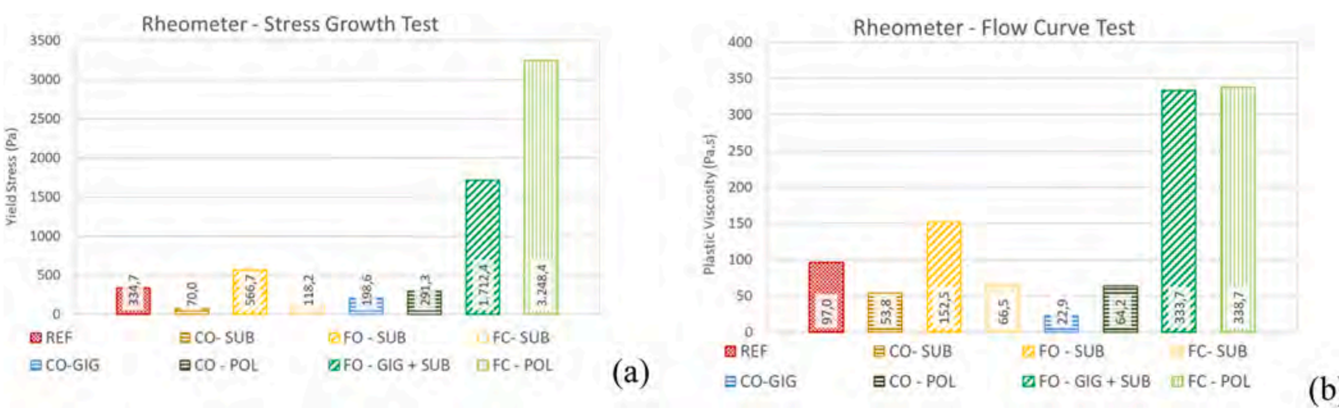


Fig. 8. Yield Stress (a) and plastic viscosity (b) of investigated concrete mixes.

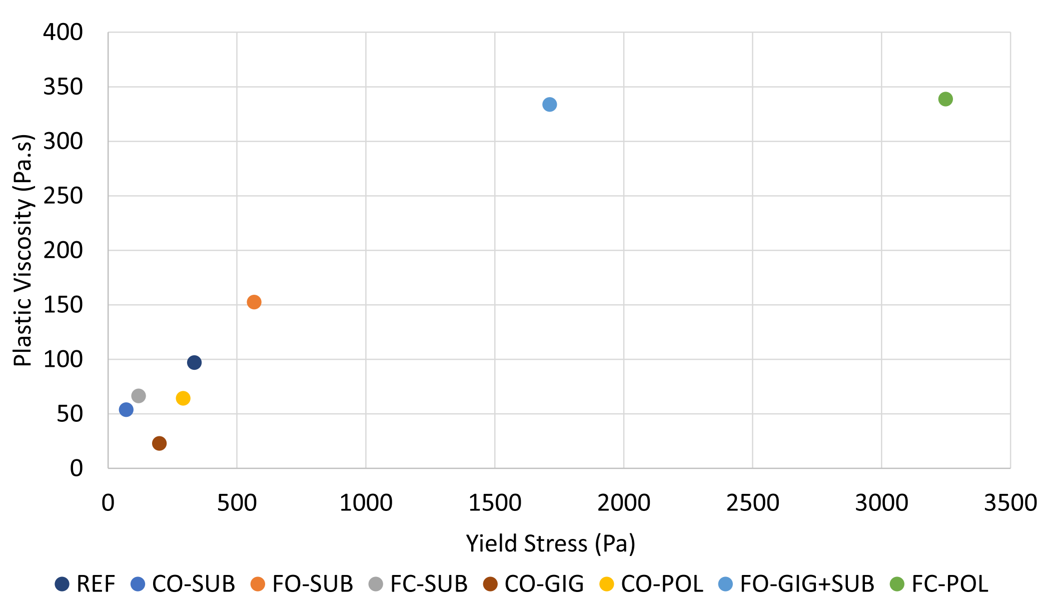


Fig. 9. Concrete workability parameters of investigated mixes.

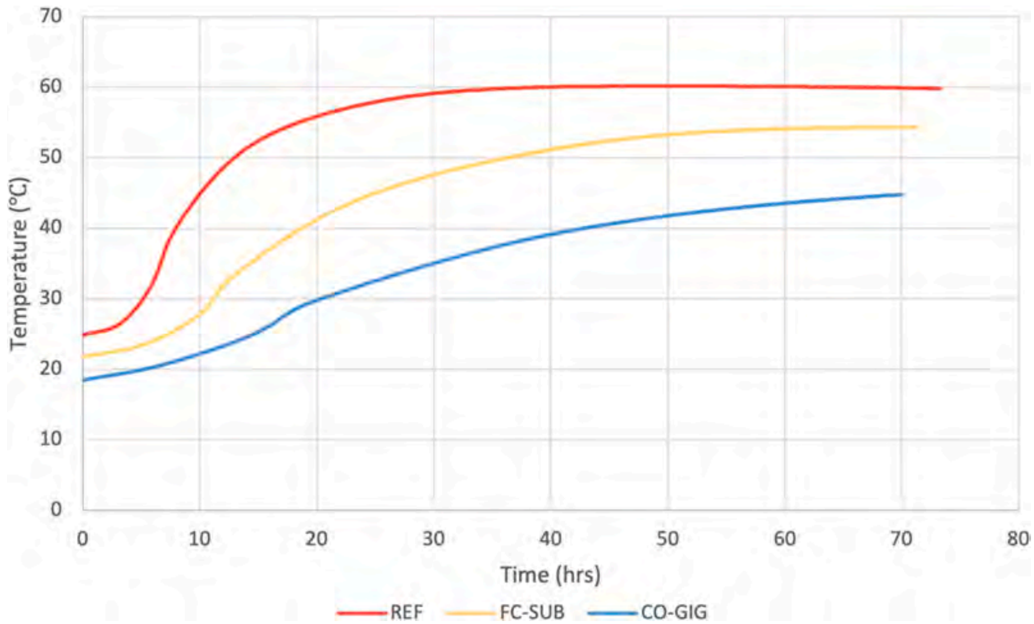


Fig. 10. Temperature (° C) of concrete hydration versus time (hours).

Table 13
Bulk density and porosity of concrete as a function of CMWGs ratio.

	C-REF	C-F10%-G30%
Porosity (%)	14.8 (6)	4.6 (0.4)

Note: Standard deviation between parentheses

GIG+SUB (natural sand replaced by fine CMWGs) and FC-POL (replacement of sand and gravel for POLTEGOR). The static yield stress (stress growth test) and the dynamic yield stress (flow curve test) of reference concrete were higher than the one corresponding to CMWGs coarse aggregates (CO mixes). The contrary occurred for the FO mixes (with fine CMWGs). This means that the reference mix was less fluid compared to CO mixes but it was more fluid than FO mixes. This fact makes sense due to the higher amount of fines in FO mixes compared to CO mixes. In fact, the replacement of only fines generally made the mix more viscous and less flowable, while the replacement of coarse aggregate had the opposite effect.

Fig. 9 summarizes the whole set of rheological properties of the investigated mixes.

•Isothermal calorimeter

An adiabatic concrete calorimeter from CONTROLS® was employed in this study to monitor the heat of hydration. The reference concrete reached the highest heat of hydration as shown in Fig. 10, whereas the replacement of natural coarse aggregate and both, fine and coarse natural aggregate with CMWGs resulted into a slower development and an overall reduction in the hydration heat. This is consistent with the retard in final setting times and in the development of hydration reactions commented above.

•Porosity of concrete

The open porosity decreased with the increase in the incorporation of CMGWs (Table 13) which agreed with mortar results shown in Section 4.1. This was due to the refinement of the pore structure caused by the presence of CMWGs, coherently with results of previously reported tests.

•Compressive strength

As previously reported in Table 8, compressive strength was determined on similar cylindrical specimens: CYU used cylinders 110 mm in diameter and 220 mm high whereas PoliMi cylinders were 100 mm in diameter x 300 mm high. The compressive strength at 28 days for the reference concrete tested at either laboratory was very similar for both

Table 14
Compressive strength values of concretes obtained from cylinders.

Specimen	Curing Time (days)	Average (MPa)	Variation (%)
C-REF (CYU)	7 days	27.3	-
C-F10%-G30% (CYU)		18.6	31.86
REF (PoliMi)	10 days	40.3	-
FO-GIG (PoliMi)		33.3	17.37
FO-SUB (PoliMi)		31.7	21.34
CO-SUB (PoliMi)		34.3	14.89
FC-GIG (PoliMi)		30.8	23.57
FC-SUB (PoliMi)		37.7	6.45
C-REF (CYU)	14 days	31.9	-
C-F10%-G30% (CYU)		24.0	24.67
REF (PoliMi)	28 days	45.8	-
FO-GIG (PoliMi)		36.7	19.93
FO-SUB (PoliMi)		33.5	26.86
FO-GIG+SUB (PoliMi)		32.8	28.38
CO-GIG (PoliMi)		30.4	33.62
CO-SUB (PoliMi)		36.4	20.52
CO-POL (PoliMi)		24.7	46.07
FC-GIG (PoliMi)		35.1	23.36
FC-POL (PoliMi)		27.3	40.39
FC-SUB (PoliMi)		39.8	13.10
C-REF (CYU)		44.6	-
C-F10%-G30% (CYU)		36.3	18.65
REF (PoliMi)	60 days*	38.7	-
FO-SUB (PoliMi)		32.7	15.5
FO-GIG+SUB (PoliMi)		30.6	20.8
CO-GIG (PoliMi)		29.8	23.0
CO-SUB (PoliMi)		33.9	12.4
CO-POL (PoliMi)		28.5	26.4
FC-SUB (PoliMi)		40.8	-5.6
REF (PoliMi)	90 days*	43.7	-
FO-SUB (PoliMi)		38.6	11.8
FO-GIG+SUB (PoliMi)		33.2	24.1
CO-SUB (PoliMi)		42.2	3.4
CO-POL (PoliMi)		28.6	34.7
FC-SUB (PoliMi)		37.3	14.8

* Values obtained from cubical specimens; the corresponding cylinder strength value was obtained by means of the equation f_c=0.83•R_c, where: f_c and R_c are the compressive strength values from cylindrical and cubic specimens, respectively.

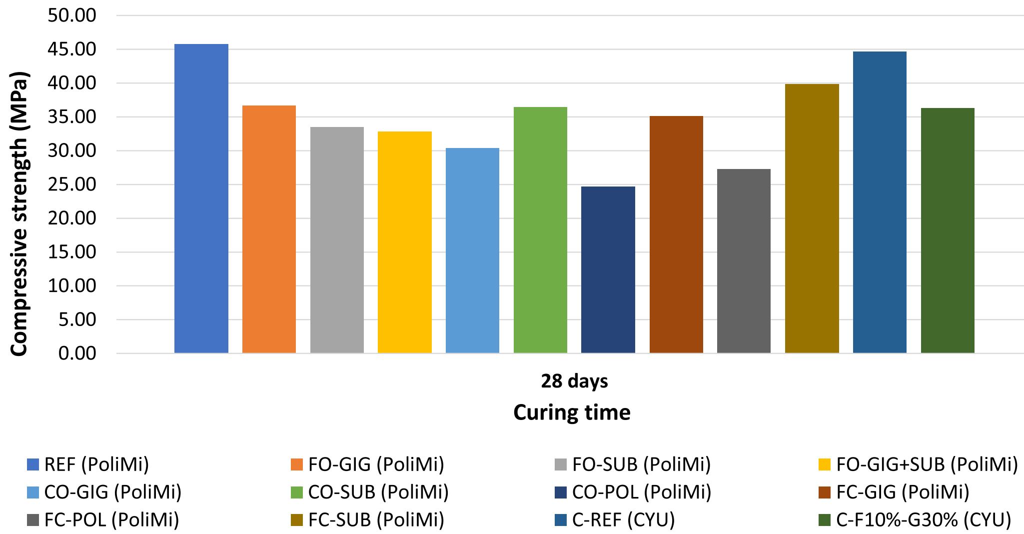


Fig. 11. Compressive strength values of concretes after 28 curing days.

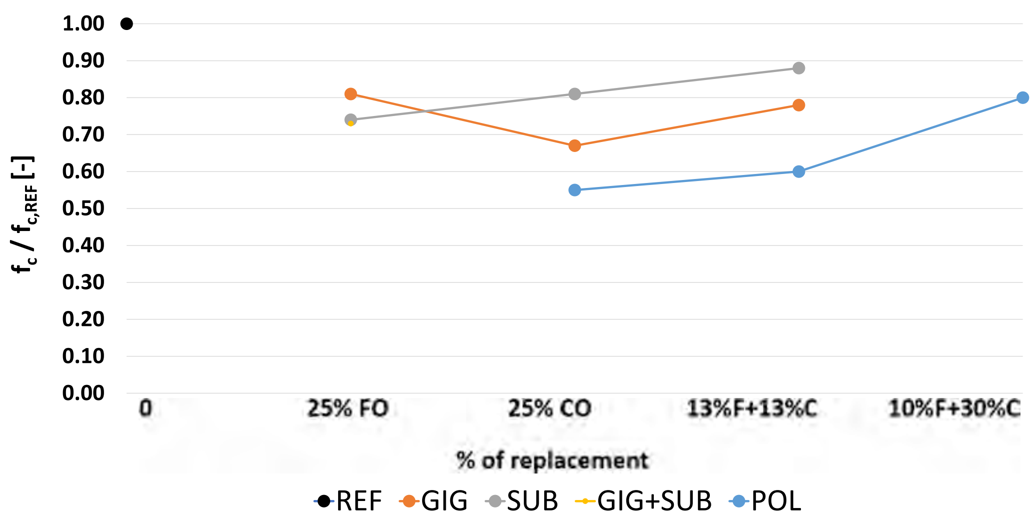


Fig. 12. Dimensionless compressive strength values at 28 days versus % of replacement.

universities, 45.8 MPa at PoliMi and 44.62 MPa at CYU (Table 14 and Fig. 11), which corroborates the significance of the present study.

PoliMi replaced 25% of natural aggregates by CMWGs (sand in the case of FO mixes, coarse in CO mixes) and 13% sand and 13% gravel in the FC mixes. The CMWGs were from different providers as indicated in Table 2. On the other hand, CYU mix was with 10% replacement of sand and 30% of gravel.

In Table 14 the compressive strength values are also reported for specimens tested at 7, 10, 14, 28, 60 and 90 days. All the values refer to cylindrical specimen although cylinders were tested at 7, 10, 14 and 28 days whereas the values at 60 and 90 days were obtained from cubical specimens; anyway the values remain substantially stable after the 28 days. In order to correctly perform the comparison, the values of the compressive strength obtained from cubic were transformed into cylinder strength using the formula: $f_c=0.83\bullet R_c$, where: f_c and R_c are the compressive strength values from cylindrical and cubic specimens, respectively.

Focusing on the results at 28 days, the replacement with sand provided by GIG (FO-GIG) resulted into the highest compressive strength value compared to the mixes using SUBTERRA fines and the combination of GIG and SUBTERRA (FO-SUB and FO-GIG+SUB respectively). On the other hand, coarse aggregates from SUBTERRA provided concretes (CO-SUB) with higher compressive strengths than those with GIG coarse aggregates (CO-GIG). Regarding the concretes with fine and coarse substitution, concrete with SUBTERRA CMWGs featured the highest compressive strength values (FC-SUB), whereas concretes with coarse aggregates from POLTEGOR provided the lowest. The different mechanical characteristics of the employed CMWGs can be deemed as responsible for the detected differences in the results.

All these conclusions can be also observed in Fig. 12 where the dimensionless compressive strength is shown versus the % replacement indicating the different CMWGs providers, interestingly confirming that a “blend” of fine and coarse CMWGs can be beneficial in achieving a better compressive strength performance than when coarse CMWGs are employed alone; this can be explained through both a better assorted grain size distribution in the granular solid skeleton as well as by a toughening effect on the coarse CMWG/mortar interface which the fine (r) fraction of the CMWGs can provide.

Finally, experimental compressive strength values along time obtained by both universities were compared with the empirical Eurocode 2 formula: $f_{cm}(t) = \beta_{cc} \times f_{cm}$ where: f_{cm} (t) is the average concrete compressive strength at an age of t days, β_{cc} is a coefficient which depends on the age of the concrete t and f_{cm} is the average compressive strength at 28 days. Fig. 13 shows the dimensionless compressive

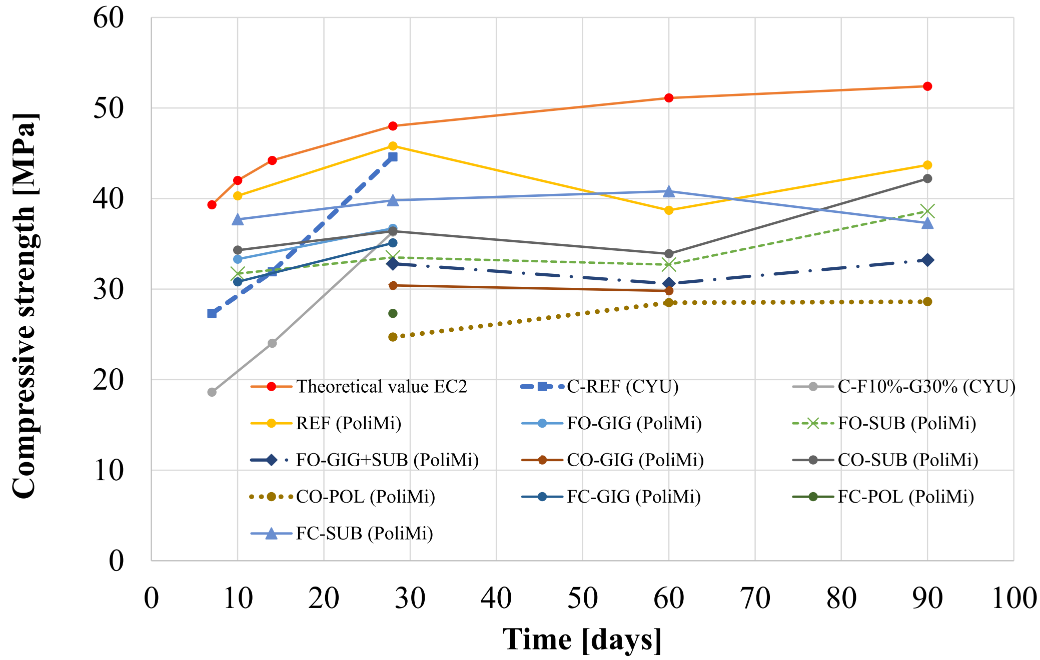


Fig. 13. Dimensionless experimental compressive strength values compared with the dimensionless theoretical compressive strength values calculated according EC2.

Table 15
Elastic modulus of investigated mixes at different investigated curing ages.

Specimen	Curing Time (days)	Average (MPa)	Variation (%)
REF (PoliMi)	10 days	31394.7	-
FO-GIG (PoliMi)		28192.3	10.20
FO-SUB (PoliMi)		33701.8	-7.35
CO-SUB (PoliMi)		33585.7	-6.98
FC-GIG (PoliMi)		26884.3	14.37
FC-SUB (PoliMi)		29868.7	4.86
REF (PoliMi)	28 days	34870.1	-
FO-GIG (PoliMi)		29295.0	15.99
FO-SUB (PoliMi)		29196.7	16.27
FO-GIG+SUB (PoliMi)		32206.1	7.64
CO-GIG (PoliMi)		24544.7	29.61
CO-SUB (PoliMi)		30514.9	12.49
CO-POL (PoliMi)		27317.6	21.66
FC-GIG (PoliMi)		28449.2	18.41
FC-POL (PoliMi)		26054.8	25.28
FC-SUB (PoliMi)		31626.3	9.30
C-REF (CYU)		46600	-
C-F10%-G30% (CYU)		33000	29.18

strength obtained dividing the fc value at each time (t) by the compressive strength value at 28 days (Fig. 13). As expectable the EC2 formula predicts well the strength development of the ordinary reference concrete, at least up to 28 days, upon which the predicted slight strength increase has not been confirmed by experimental results. For concretes containing CMWGs as recycled aggregates the strength development trend seems reasonably matched, though, provided the formula is calibrated on actual 28 days strength values. Moreover, some compressive strength values regarding CMWGs at ages longer than 28 days were higher than the unit which means that higher values were obtained, however for most concretes with CMWGs the values were lower than the unit for ages longer than 28 days as shown in Fig. 13.

4.4. Elastic modulus

The results summarized in Table 15 and Fig. 14 show that the elastic modulus decreased when natural aggregates were replaced with CMWGs in the percentages indicated in Table 15, which range from 7.64% (FO-GIG+SUB) to 29.61% (CO-GIG). When only fine aggregates or both fine and coarse aggregates were replaced by CMWGs, except for few exceptions, the decrease in the elastic modulus (7.64–16.27%) was lower as compared to the replacement of natural fine aggregates only (25%). Results are coherent with trends already detected for the compressive strength and reasonably attributable to the different characteristics of the employed CMWGs and to their interaction with the binding phase, as it will be explained after through thin section analysis.

On the other hand, when replacement was with coarse aggregates, the elastic modulus decreased more (21.66–29.61%) as compared to the replacement of fine aggregates and the decrease was similar to the rate of aggregate replacement (25%). The only exception was when the replacement was carried out with CMWGs from SUBTERRA, in this case the reduction was lower (12.49%) as shown in Table 15.

Finally, Fig. 15 shows the comparison between the experimentally measured values of the Yoing modulus and the ones obtained from the Eurocode 2 formula, which proved fairly accurate:

$$E_{cm}(MPa) = 22 \times [(f_{cm} in\ MPa)/10]^{0.3} \times 1000.$$

4.5. Splitting tensile strength

CYU determined the splitting tensile strength of their concretes (reference concrete and C-F10%-G30%). The results are shown in Fig. 16 and Table 16. As expected, the splitting strength decreased when aggregates were replaced with CMWGs. The decrease was more evident when concretes reached the 28 days of curing probably due to the lower strength of aggregates which have lower effectiveness in playing a crack arrestor role, also because of the interface strengthening with age and toughening due to the effects of CMWGs fines. The rates of decrease in splitting strength compared to the reference mix were similar to the percentage of coarse aggregate (30%) being 27.66%, 28.83% and 36.17% at 7, 14 and 28 days, respectively.

4.6. Flexural strength and fracture energy

The flexural strength of concretes was evaluated by PoliMi. Fig. 17

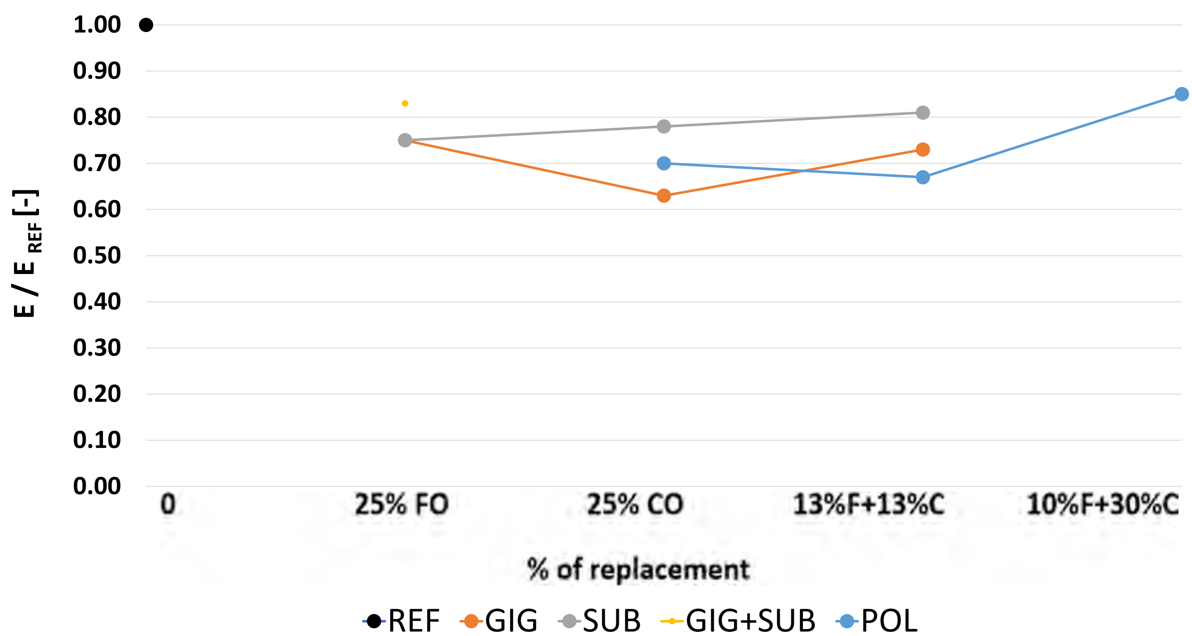


Fig. 14. Dimensionless elastic modulus of concretes at 28 days versus % of replacement.

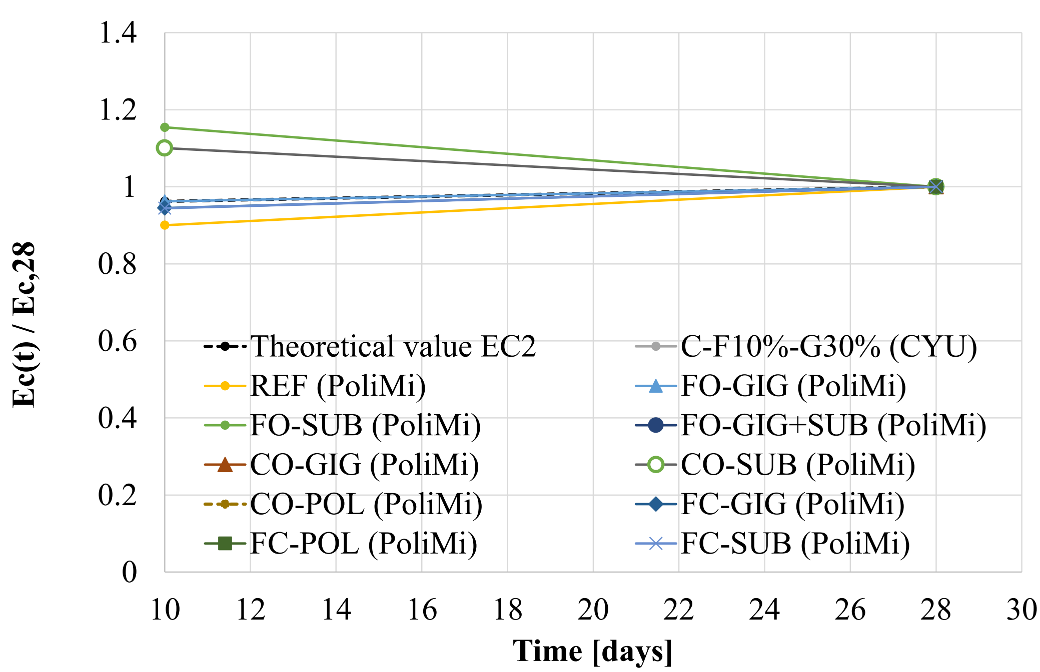


Fig. 15. Dimensionless experimental elastic modulus values compared with the dimensionless theoretical value calculated according to Eurocode 2.

shows the average flexural strength values for the tested concretes after 28 curing days. After 28 days, the mixes with CMWGs reached lower flexural strength values compared to the reference concrete. No significant differences were observed among CMWGs aggregates from the different providers. In conclusion, although the presence of fine CMWGs provides a densification of the mortar and the coarse aggregates perform a crack arrestor role, no significant differences due to the size of the aggregate (sand or coarse) were observed, these positive effects being likely jeopardized by the weakness of CMWGs (as confirmed by the fracture cross section image in the inset in Fig. 17 which, besides a homogenous distribution of the coarse CMWGs aggregates, clearly highlights the complete and net breakage of the same aggregates upon flexural failure of the specimen), contrarily to what happened to natural aggregates, some of whose particles can be clearly seen intact protruding from the fracture plane.

In addition to flexural strength, the flexural tests, performed in displacement control and with the measurement of the crack opening displacement, allowed to calculate the fracture energy, as the area under the load vs crack opening displacement curve and divided by the area of the ligament cross section (Fig. 18). The mixes with replacement of natural coarse aggregates by coarse CMWGs reached the highest values of fracture energy while the mixes with replacement of fine aggregates showed a fracture energy similar to the energy of reference concrete. The energy dissipated for the mixes with CMWGs was equal or sometimes higher than the value calculated for reference concrete, confirming the findings and explanations exposed above with reference to

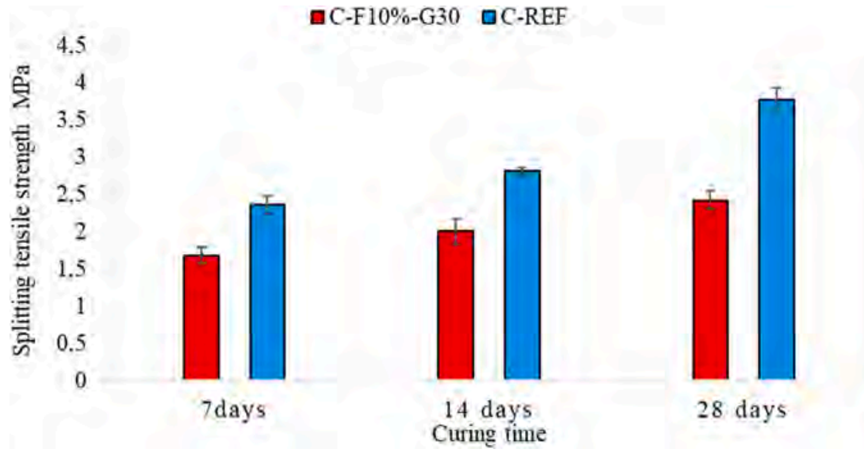


Fig. 16. Splitting tensile strength as function of natural aggregate replacement.

Table 16
Splitting tensile strength of concrete at different curing age.

Curing age	C-REF	C-F10%-G30%
7days	2.35 (0.12)	1.7 (0.11)
14 days	2.81 (0.04)	2 (0.16)
28 days	3.76 (0.16)	2.4 (0.12)

Note: Standard deviation between parentheses

flexural strength.

4.7. Shrinkage

Shrinkage was measured by PoliMi after 1, 2, 3, 7, 14, 21, 28, 60 and 90 days on prismatic specimens 100×100×500mm (Fig. 19). At 90 days, the reference mix showed the lowest shrinkage. All the mixes containing CMWGs, irrespective of the size assortment and of the provenance featured a quite higher shrinkage deformation as compared to the reference mix, surely due to the presence of coarse aggregate in the concrete mix, which, being more porous, absorbs more water and enhances the pathways moisture exchange between the specimen and the surrounding environment.

4.8. Mineralogical and microtextural characterization

The analysis was performed on four CMWG concrete samples, produced at PoliMi, provided by GIG (CO-GIG), POLTEGOR (CO-POL) and SUBTERRA (CO-SUB and FC-SUB), plus the reference one. The CWGMs used in the samples from Poland correspond mainly to argillitic grains rich in kaolinite, illite and chlorite often containing laminations of coal (Fig. 20A), and framboidal pyrite (Fig. 20B). Quartz and altered K-feldspar and plagioclase are the main components of the CWGMs in the samples from SUBTERRA In these two samples, coal and pyrite, generally with a subhedral shape, are less abundant than in the Polish materials. Besides the CWGMs used as coarse aggregate, the mineralogical characterization confirmed that the gravel and sand particles are mainly composed of calcite, dolomite, quartz and K-feldspar. The mineralogical characterization also revealed that in samples CO-POL, CO-GIG, CO-SUB and FC-SUB, the cement paste is composed mainly of calcium silicate hydrate (C-S-H) and calcium aluminum silicate hydrate (C-A-S-H), being the latter the prevalent hydrated phase in CO-POL, as can be observed in the respective false colour mineral maps (Fig. 21). Note that the QEMSCAN® is not able to distinguish between hydrated and non-hydrated phases and the minerals list in Fig. 21 therefore comprises both (e.g. calcium silicates corresponds both to unhydrated calcium

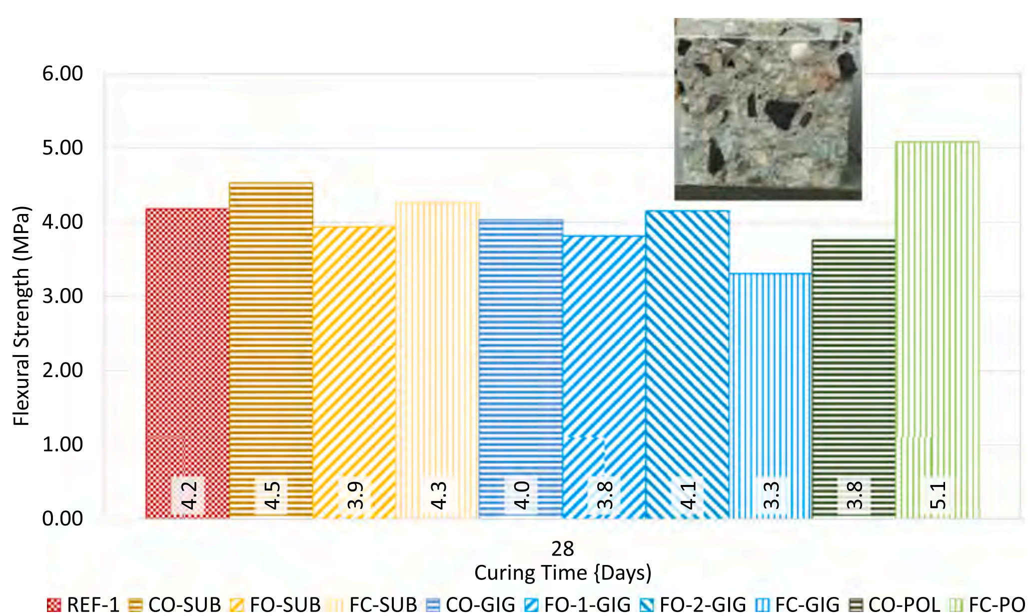


Fig. 17. Flexural strength vs curing days (the inset shows the fracture plane of the specimen after the flexural failure test).

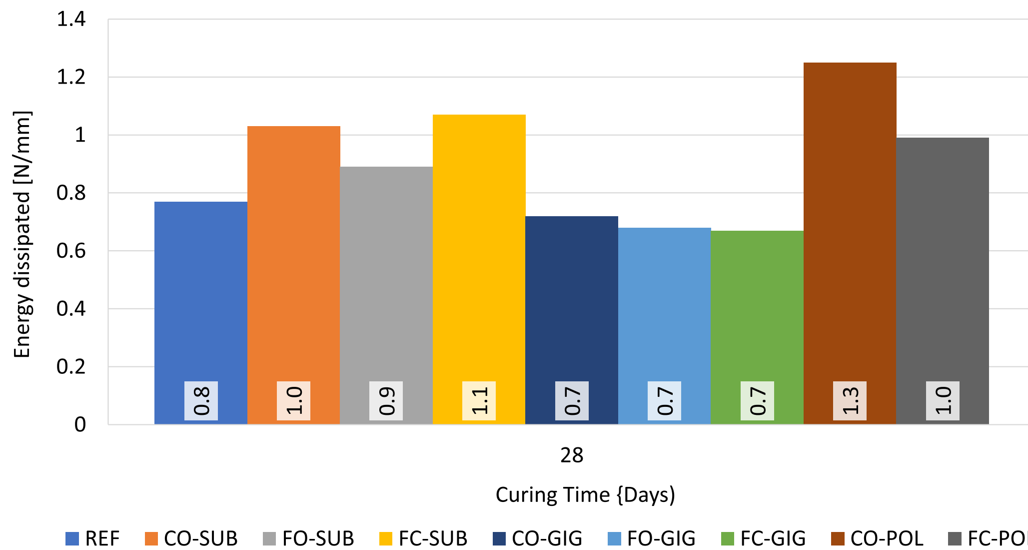


Fig. 18. Energy dissipated vs curing days.

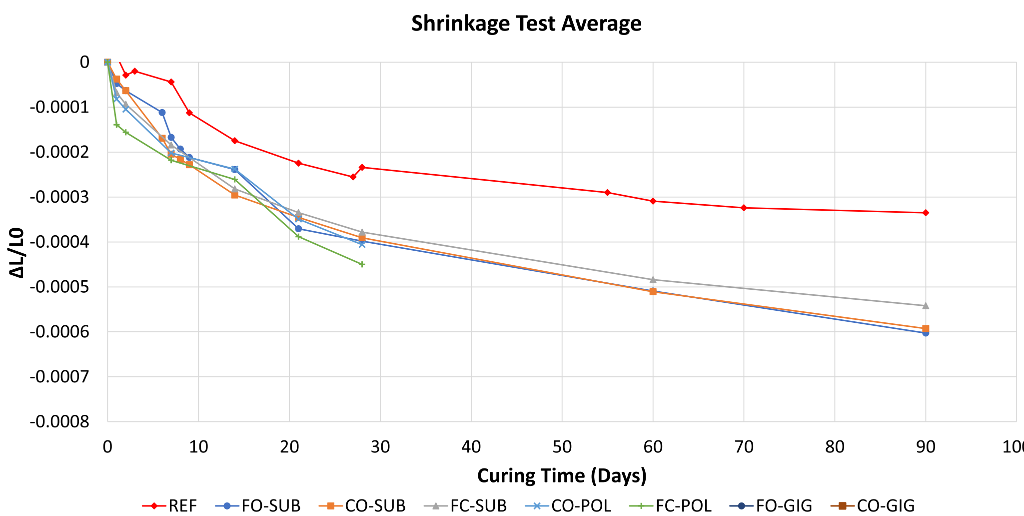


Fig. 19. Shrinkage results for concrete mixes.

silicate phase and to calcium silicate hydrate (C-S-H)). Taking into account that the only difference in the formulation of the concrete samples resides in the nature/composition of the used CWGMs, a possible explanation for the distinct hydration phases could be given by differences in the content and/or grading of fines and extra fines (dusts) that could be acting as pozzolans especially in CO-POL. A detailed study of the mineralogical and microtextural aspects of the aggregates included in all the formulations used in this investigation should be carried out in order to more accurately understand the behaviour of mortar and concrete using CWGMs.

Under the petrographic microscope, and in agreement with what was previously referred in respect to the shrinkage of concrete using coarse aggregates from CWGMs, shrinkage microcracks were observed cross-cutting the cement paste and skirting coarse aggregates particles especially in CO-POL and CO-GIG samples (Fig. 20C and F). Although shrinkage microcracks are also present in the cement paste of the REF sample, these are less abundant. Several CWGMs coarse particles in the CO-GIG and CO-POL samples showed evidence of internal cracking due to the self-contraction of clay during desorption (Fig. 20D). The same was observed for the samples using Spanish CMGMs (Fig. 20f), but more scarcely. It should also be noted that sample CO-SUB presents a higher amount of entrapped porosity, as can be observed in Fig. 20G and 21D.

All these anisotropies can contribute to support the weaker mechanical behaviour found for concrete using the concerned geomaterials. Furthermore, overall, the samples containing the Spanish CWGMs show a better mechanical performance in comparison to the samples containing Polish waste, which may be justified by their lower content in clay. According to several researchers (e.g., Kawabata et al., 2021 [28], Zhao et al., 2022 [29] and references therein), clay content positively correlates with drying-shrinkage of aggregate/concrete when the used aggregate has abundant clay minerals. In spite of the lower matrix porosity in samples using CWGMs, the presence of a patchy carbonation (Fig. 20H) due to a higher porosity of the coarse aggregate particles from CMWGs and to the cracking from shrinkage is well visible in CO-GIG and CO-POL samples.

5. Conclusions and recommendations

The results have been reported and analyzed in this paper of a comprehensive experimental program to evaluate the feasibility of replacing natural aggregates with Coal Mine Waste Geomaterials (CMWGs) in concrete and the influence of the percentage of replacement on the properties of structural concrete.

The main outcomes of this study can be summarized as follows:

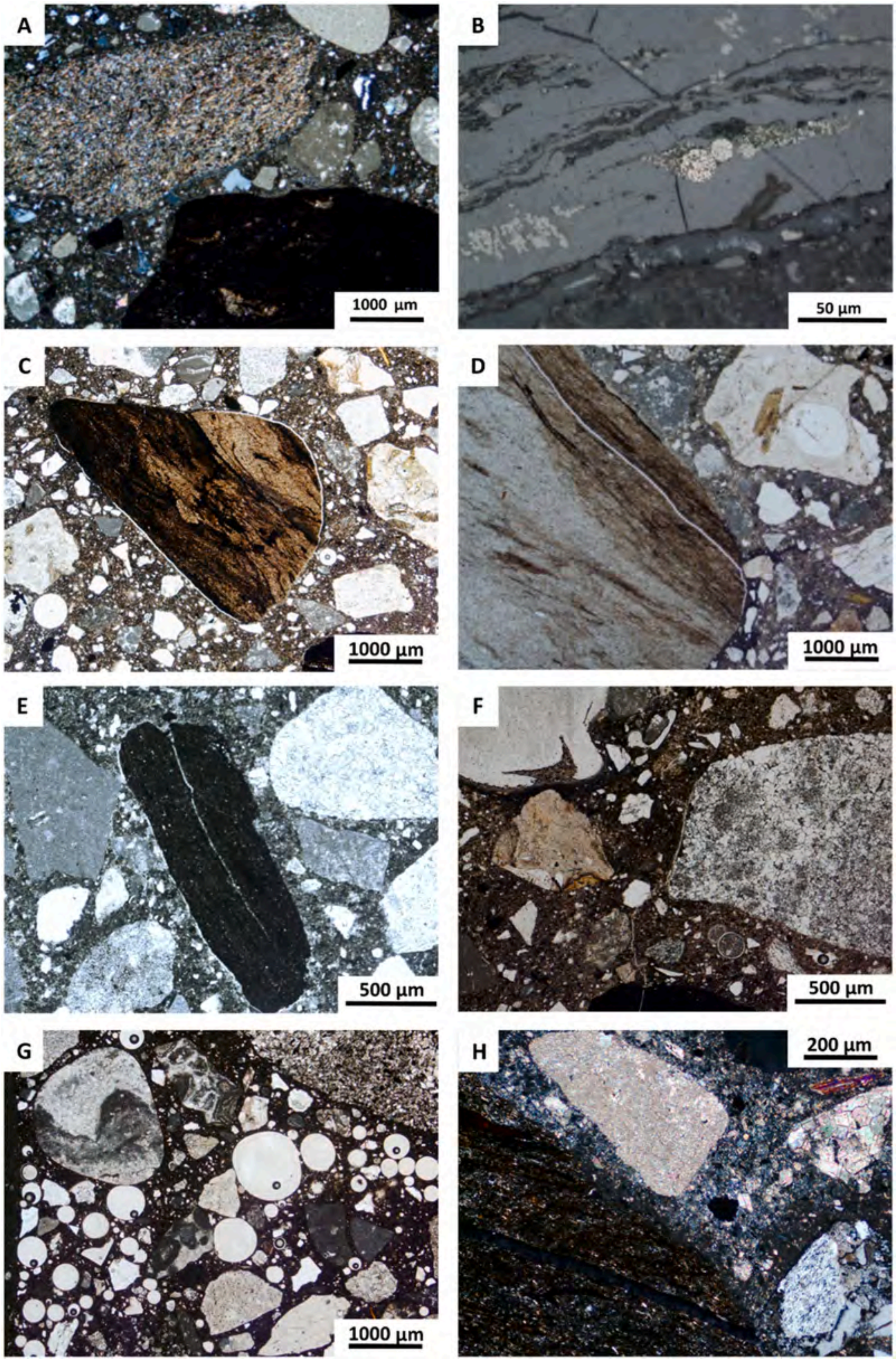


Fig. 20. Microphotographs of CO-POL (A to C and F), CO-GIG ((D and H), CO-SUB (G) and FC-SUB (E) samples. Photos A and H in cross-polarized light. Photos C to G in plane polarized light.Photo B in reflected light. A) Argilitic CWGMs coarse aggregate particles; B) Framboidal pyrite; C) Shrinkage microcracks skirting a clay-rich CWGM coarse particle; D and E) Internal microcracks in clay-rich CWGMs; F) Shrinkage microcracks in the cement paste; G) Abundant entrapped air voids in the cement paste; H) Patchy carbonated areas in the cement paste (lighter areas).

- The replacement of natural sand by fine CMWGs did not significantly affect the workability of the mortars;
- Rheometer tests on concrete showed that the static yield stress (stress growth test) and the dynamic yield stress (flow curve test) of reference concrete were higher than the one of mixes with CMWGs replacing coarse aggregates (CO) mixes. The contrary occurred for the FO mixes (with fine CMWGs). This fact means that the reference mix was less fluid compared to CO mixes but it was more fluid than

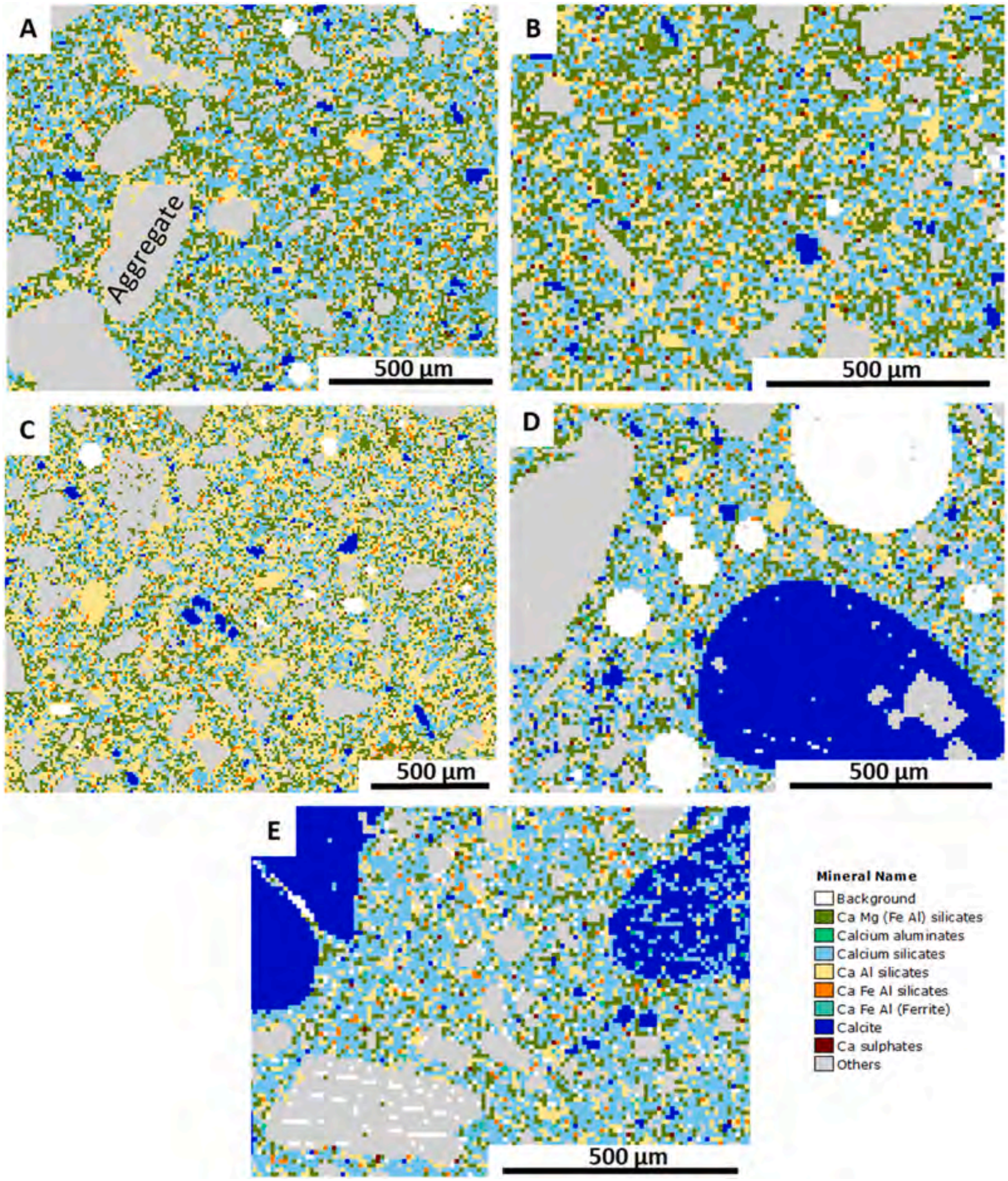


Fig. 21. Selected regions of QEMSCAN® false colour mineral maps focusing on the cement paste phases from samples REF (A), CO-GIG (B), CO-POL (C), CO-SUB (D) and FC-SUB (E) showing calcium silicate hydrate (C-S-H) as the main constituent of the cement paste in all samples, except in CO-POL where calcium aluminum silicate hydrate (C-A-S-H) is the main binder phase.

FO mixes. This fact has sense due to the higher amount of fines in FO mixes compared to CO mixes. In fact, the replacement of only fines generally made the mix more viscous and less flowable while the replacement of coarse aggregate had the opposite effect;

- The calorimeter allowed to determine the heat of hydration of the concrete mixes. The reference concrete reached the highest heat of hydration, whereas the replacement of natural coarse aggregate and both, fine and coarse natural aggregate with CMWGs resulted positive since the hydration heat diminished;
- The presence of CMWGs reduces the porosity of mortars and concretes since CMWG aggregates have a significant role not only in filling the concrete pores but also refining the pore structure which can be explained due to the fact that CMWGs could help to reduce porosity through pozzolanic reactions;
- The presence of CMWGs only slightly affected the shrinkage of cement mortars. On the contrary, the shrinkage of concretes was higher for the mixes with CMWGs. The higher porosity of coarse aggregates obtained from CMWGs increased the water absorption and hence increasing the shrinkage of concretes. This seems to be confirmed by petrographic analyses, which revealed microcracks in the CMWG coarse particles as well, as effect of the desorption. In the mortar this effect was mitigated due to the absence of coarse aggregate which this high porosity in case of CMWGs have a significant effect on the water absorption and exchange with the surrounding environment and hence on the shrinkage;
- The mechanical properties of mortars and concretes (compressive and flexural strength) decreased when aggregates were replaced with CMWGs. The decrease was usually more evident when concretes reached the 28 days of curing. The compressive strength decreased as much as the CMWGs percentage increased in mortar and concrete mixes being the percentage of variation with respect to the reference mix similar to the percentage of replacement of aggregates;
- The elastic modulus decreased when natural aggregates were replaced with CMWGs. In mortars, the decrease was slight whereas

was more evident in concretes when coarse aggregates were replaced by CMWGs;

- The splitting tensile strength decreased when natural aggregates were replaced with CMWGs. The decrease was more evident when concretes reached the 28 days of curing probably due to the lower strength of aggregates which have lower effectiveness in playing a crack arrestor role, despite the densification and toughening of the interface transition zone due to the CMWGs fines;
- The mixes with replacement of natural coarse aggregates by coarse CMWGs reached the highest values of fracture energy while the mixes with replacement of fine aggregates showed a fracture energy similar to the energy of reference concretes. In fact, concrete mixes with CMWGs reached lower flexural strength values compared to reference concrete whereas concrete mixes with CMWGs reached similar or even higher energy dissipated values compared to reference;
- The preliminary assessment regarding the mineralogical and microtextural characterization clearly supports the results from the mechanical tests.

As a whole, the replacement of natural aggregates by CMWGs has resulted an interesting and sustainable option for civil engineering applications in which a lower performance from the point of view of mechanical performance may not be a problem for its functionality.

CRediT authorship contribution statement

Oumayma Aboutaybi: Investigation, Formal analysis, Data curation. **Elhem Ghorbel:** Writing – review & editing, Resources, Project administration, Methodology, Conceptualization. **Jens Andersen:** Investigation, Data curation. **William Nash:** Investigation, Data curation. **Marco Del Galdo:** Methodology, Investigation, Data curation. **Gavyn Rollinson:** Methodology, Investigation. **Violeta Ramos:** Writing – original draft, Methodology, Investigation, Data curation. **Estefania Cuenca:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Rich Crane:** Writing – review & editing, Project administration, Methodology, Funding acquisition, Formal analysis, Conceptualization. **Liberato Ferrara:** Writing – review & editing, Validation, Supervision, Project administration, Methodology, Funding acquisition, Formal analysis, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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