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Concrete-Polymer Composites in Circular Economy

Proceedings of the 17th International Congress on Polymers in Concrete (ICPIC 2023)

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Preface

The 17th International Congress on Polymers in Concrete has taken place in Warsaw, Poland, on September 17–20th, 2023. It was organized by the International Congress on Polymers in Concrete, the Warsaw University of Technology and the Building Research Institute (ITB) under patronage of ACI, RILEM, the Rector of Warsaw University of Technology, Polish Association of Civil Engineers, the Section for Building Materials Engineering of the Committee on Civil Engineering of the Polish Academy of Sciences and Polish Association of Civil Engineers and Technicians.

The ICPIC congresses are cyclic events organized since 1975 by the International Congress on Polymers in Concrete. This non-profit organization gathers specialists in research and the practical use of polymers in concrete.

The aim of the 17th ICPIC 2023 was to create a forum for the exchange of multidisciplinary knowledge referring to the application of polymers "in concrete" and "on concrete": from the modification of concrete composition with modern admixtures and additives, through alternative binders (like geopolymers, sulphur concrete, and others), polymer composites for reinforcing concrete (including fibres, FRP bars and FRP strengthening systems), improvement of concrete surface properties (by impregnation, hydrophilization and coatings) to unique properties like self-healing, self-cleaning or energy consumption control with PCM (Phase Changing Materials).

The main issues of the 17th ICPIC held at Warsaw were challenges in the field of Concrete-Polymer Composites (C-PC), concerning the implementation of a circular economy. Therefore, an essential part of the 17th ICPIC 2023 was the Special Symposium entitled "C-PC in Circular Economy: Searching for a New Paradigm", organized by the Building Research Institute (ITB) under the chairmanship of Professor Lech Czarnecki.

The Part I of the proceedings contains the papers deal with the main topics of the Special Symposium. Part II of the proceedings was divided into seven chapters containing the papers deal with issues related to application and challenges for c-pc in circular economy, alternative binders, admixtures and additives, repair and protection of concrete structures, reinforcement and strengthening, improvement of the C-PC properties, special properties of concrete. At the 17th ICPIC Congress the papers have been presented by authors from 19 countries. We are very grateful to all authors for their participation, especially for exciting and fruitful discussions during the 17th ICPIC Congress on the new paradigm for Concrete-Polymer Composites, which will meet the challenges of circular economy implementation. The content of this proceedings clearly indicates a necessity for such discussion. I hope that the papers presented in the proceedings will be a source of inspiration and a guide for new ideas that have been outlined during the 17th ICPIC in Warsaw in the field of sustainable concrete-polymer composites.

On behalf of the Organizing Committee

Andrzej Garbacz
President of the International Congress on
Polymers in Concrete

Optimization of Eco-Sustainable, Form-Stable Phase Change Material to Be Incorporated in Aerial-Lime-Based Mortars

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Abstract. The building sector faces a challenge to find innovative and sustainable ways to increment the energy-efficiency of buildings and reduce their environmental impact. Recently, the incorporation of phase change material (PCM), based on a polymeric active phase (PEG-1000) in waste stone aggregates, has proven to be a promising option to be used for building restoration. Mortars that include PCM aggregates demonstrated to have favorable thermal properties, that would lead to a reduction of energy requirement for heating/cooling needs. However, the inclusion of aggregates impregnated by PEG causes a reduction in the mechanical properties of the mortars possibly due to (i) a lack of compatibility between aggregate and binder, or (ii) a problem with the confinement of the PEG, causing its dispersion in the mortar. Therefore, the aim of this study was to investigate the causes associated to the reduction of the mechanical properties and propose a method to prevent it. Preliminary results showed that, given its high water solubility, the PEG 1000 included in the stone aggregates tends to be *washed away* when these aggregates are incorporated in the mortar mixture. This hypothesis was confirmed by FTIR spectroscopy. Therefore, an additional confinement method using a layer to coat the stone aggregates impregnated by PEG 1000 was proposed in this study. Different materials were tested as coating layer: powder calcium hydroxide, milk of lime (suspension of $Ca(OH)_2$ in water), pozzolana, and cocciopesto. Carbonated mortar samples using the proposed coated aggregates were, then, analyzed using FTIR to evaluate the efficiency of this encapsulation methodology. Preliminary results suggested a relevant improvement in terms of PEG confinement.

Keywords: Phase change materials (PCMs) · Lime-based mortars · Thermal energy storage (TES) · Circular economy

1 Introduction

The environmental impact of human activities is a concerning issue that has been tackled in recent years by many disciplines and industrial sectors. Particularly for the construction industry, the energetic consumption of a building during its use is considerably

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higher than the energy required for its construction [1, 2]. This large consumption is mainly due to cooling and heating systems [3]. Thus, is fundamental to mitigate this issue by finding solutions to improve the energy efficiency of buildings. On the other hand, the renovation and restoration of historical buildings to prolong their usable life has also a positive impact on the environment. It has been estimated that the renovation of a building could save 30–60% of energy with respect to constructing a brand-new one [4]. The renovation of historical buildings also permits to implement innovative solutions to improve the building's energy consumption, for example Phase Change Materials (PCM) for passive thermal regulation.

The implementation of PCM in the mortar mixture has been proposed in the last decade [3, 5] as a suitable solution to obtain better thermal stability in the buildings. These materials are characterized by a large latent melting heat; thus, they are able to absorb/release thermal energy trough changing its physical phase [6]. PCM can act, in fact, as passive systems reducing the total energy demand required for the maintenance of comfort conditions inside a building. Even if their implementation in lime-based mortars is recent [7–9], the results show a promising improvement of the thermal properties of the PCM-based mortars. A sustainable solution was employed for the incorporation of the polymeric phase of the PCM, i.e. PEG 1000 (Poly(ethylene glycol)), in the mortars. Form-Stable method was employed using waste pieces of Lecce Stone (LS) as the support matrix for the form stable PCM (FS-PCM) [7, 10–12]. The inclusion of these aggregates, on the other hand, causes a reduction in the mechanical properties of the mortars, forcing to reformulate the mix-design in order to compensate for the loss in mechanical properties [11].

It was hypothesized that the reduction in mechanical properties may be due to (i) a lack of compatibility between aggregate and binder, or (ii) a problem with the confinement of the PEG, causing its dispersion in the mortar. Starting from these considerations, the aim of the present work was to investigate the causes associated to the reductions in mechanical properties of the PCM-based mortars and propose some new methods to prevent it.

2 Materials and Methods

2.1 Modification of the Form-Stable Phase Change Material Aggregates

The Form-Stable Phase Change Material (FS-PCM), were obtained according to the process described by Frigione et al. [7]. These composite materials were produced using Lecce stone (LS) as the matrix for supporting the PCM. The LS is composed mainly by $CaCO_3$ and was obtained from the waste of the stone cutting process from a quarry located near Lecce (Italy). The active phase for the PCM was Poly(ethylene glycol) (trade name PEG 1000) supplied by Sigma—Aldrich (Germany). This is a non-toxic, low environmental impact, low-flammable, and relatively cheap thermoplastic polymer, with a melting temperature in the range 37°–40 °C. The composite made with LS and the PEG 1000 was the FS-PCM used as aggregate in the mortar mixture. In the present work, these aggregates were modified in four different ways, indicated as: size reduction, polishing, washing, and coating.

The size reduction was performed by mechanically compressing the FS-PCM in a mortar without grinding them. For the polishing method, sandpaper of 600 to 2500 grade was used to partially remove the outermost layer of the PEG polymer composing the PCM. Water washing with deionized water was performed in two different configurations: static and manually stirred. Finally, the application of a coating was chosen in order to obtain an external layer on the composite PCM aggregates. FS-PCM was placed in a heating plate under constant stirring (500–700 rpm) at 34ºC (i.e. at a temperature within the range of phase change of the PEG 1000). The coating material was added in an excess of approximately 20% with respect to the weight of the aggregates and mixed for 20–30 min. The coating materials were: powder calcium hydroxide (CH), milk of lime (suspension of $Ca(OH)_2$ in water) (ML), pozzolana (P), and cocciopesto (C). The latter being a fine powder made of clay materials mixed with lime and sand. The calcium hydroxide was provided by Fassa Bortolo Srl (Italy), the milk of lime was produced using the same calcium hydroxide. The pozzolana and cocciopesto were supplied by CTS company (Italy).

2.2 Composition of Mortar Formulations

With the aim of understanding the behavior of the coated aggregates when they are included in a mortar, a set of mortars were produced and subsequently analyzed trough FTIR. All mortars, possessing the same formulation, were based on aerial lime, provided by Fassa Bortolo Srl, and carbonatic sand of different granulometries, provided by the same company. To these mortars the modified aggregates were added. The composition of the mortars are reported in Table 1. The amount of water added to the mix corresponded to 36% by weight of total dry components (i.e. sum of all the weights added in the proportions expressed in Table 1).

Table 1. Proportions of dry components of the mix-design expressed in percentage by weight.

Binder: Hydrated lime ($Ca(OH)_2$)	FS-PCM or modified aggregates (CH, ML, P, C)	Sand (0.1 – 0.6mm)	Sand (0.6 – 1.4mm)	Total of dry components
30	40	10	20	**100**

2.3 Material Characterization

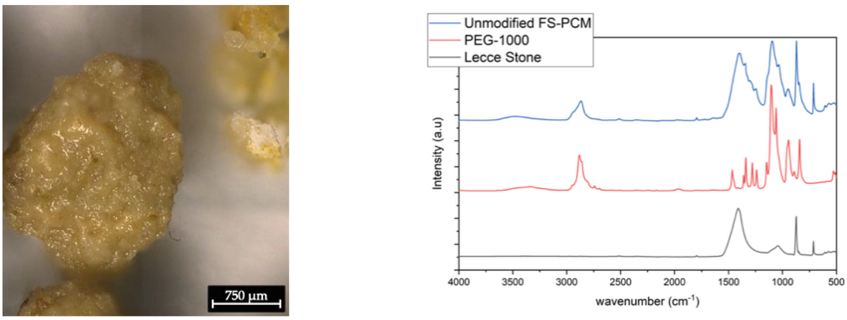
In order to characterize the surface properties of the aggregates before modification, optical microscopy observations were carried out using a Leica DM6 microscope at 5X and 10X magnifications, coupled with a Flexcam C1 camera.

Fourier-Transform Infrared Spectroscopy (FTIR) measurements were made to determine and quantify the presence of PEG 1000 in the unmodified aggregates. The same technique was also used to analyze the presence of PEG 1000 in the mortar mixture and evaluate the efficacy of the confinement method. All measurements were made using a Macro Attenuated Total Reflectance FTIR (Macro-ATR-FTIR) Thermo Nicolet iS20 spectrometer, equipped with a DTGS detector and a Smart itX diamond accessory for ATR, in the spectral range 4000 - 400 cm^{-1}, collecting 64 scans for each measurement with a 4 cm^{-1} spectral resolution (diameter of the window 2 mm).

3 Results and Discussion

The unmodified FS-PCM was characterized by a *waxy* surface clearly visible under the microscope (Fig. 1a), which indicates that PEG polymer was able to fill the porosity present in the Lecce Stone aggregates but it is also present on their surface. This observation confirmed that the reduction in mechanical properties of the PCM-based mortars can be due to a lack of compatibility between the binder and the polymer present on the aggregate, thus surface modifications might help to avoid this issue. Moreover, the average size of the aggregates is 2mm [7]. It is known that to have adequate mechanical properties, the granulometry curve of the aggregates must be compact and even [13]: as the FS-PCM aggregates are quite large, a reduction of their size could be a possible means to improve the mechanical characteristics of mortars, without modifying the PCM content.

The chemical nature of the composite aggregates was characterized by FTIR: the characteristic signals of both $CaCO_3$ stone and PEG 1000 are visible in the spectra shown in Fig. 1b. As some of the characteristic peaks of both PEG 1000 and $CaCO_3$ superpose, especially in the region of 1500−600 cm^{-1}, the identification of the presence of PEG polymer in the composite PCM was obtained through the observation of the peak at 2885 cm^{-1}.



a b

Fig. 1. (a) Optical microscopy image of a single unmodified FS-PCM aggregate and (b) FTIR spectra of the FS-PCM and of its components.

The other three modification methods implemented, i.e. polishing, water washing, and coating. Aimed at increasing the compatibility between the PCM aggregates and the mortars, by eliminating the superficial layer of PEG 1000 or by covering it.

Both size reduction and polishing modifications gave unsatisfactory results due to the reduced mechanical resistance of the impregnated Lecce Stone: when the PCM composite aggregates were either pressed or polished, they immediately shattered. The water washing method revealed that, due to the high solubility of PEG 1000 in water (value reported in product datasheet: 1500g in 30mL of H_2O), after 30 to 45 min of washing in static conditions, no PEG polymer remained in the PCM aggregates, confirmed by FTIR measurements (Fig. 2). The washing process carried out in stirring conditions was able to accelerate the *washing out* of PEG 1000, and almost no FTIR signal of PEG 1000 was found after a 5 min process. These results led to hypothesize that, when the FS-PCM aggregates are included in a mortar, part of PEG 1000 can be dispersed in it, possibly worsening its mechanical properties.

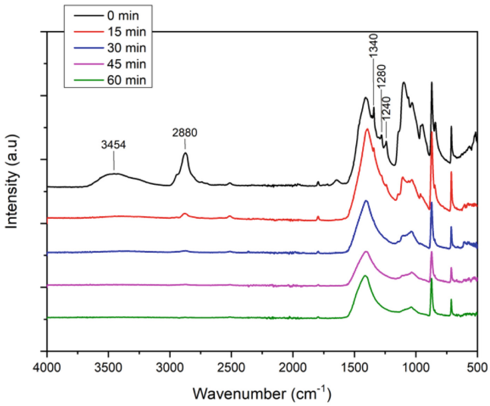


Fig. 2. FTIR spectra of the washing process under static conditions of the FS-PCM aggregates.

The coating methodology involved the application of an external layer of different materials (i.e. powder calcium hydroxide (CH), milk of lime (suspension of $Ca(OH)_2$ in water) (ML), pozzolana (P), and cocciopesto (C)) on the PCM-based aggregates, with the aim of improving the compatibility between the aggregate and the binder. In the case of the coating materials in powder form (i.e. CH, P, and C), the adhesion between the powder and the aggregate was achieved through the heating of the aggregates at a mild temperature (34 °C) in order to improve the adherence of the coating powder. The ML coating was applied at room temperature: once the aggregates were completely covered, they were left to dry for a few hours. The resulting materials are shown in Fig. 3. It is possible to observe that the color of the original FS-PCM (Fig. 3a) was changed to white by the application of both CH and LM coatings (Figs. 3b and 3c, respectively); on the other hand, the pozzolana and cocciopesto coatings modified the color into an *earth* or *brick* tone (Figs. 3d and 3e, respectively). This is an important aspect as color compatibility is an important parameter to take into account in restoration applications.

In order to evaluate the efficacy of the coating method with the different materials on the confinement of the PEG 1000 inside the stone aggregates, both FS-PCM unmodified and the modified aggregates were incorporated in a mortar mixture. The FTIR-ATR spectra obtained on the surface and on a cut section of each mortar are shown in Fig. 4. It is important to highlight that the powder analyzed was obtained by scratching the binder and carefully avoiding the aggregates. The peak corresponding to the C-H stretching of the PEG 1000 chain ($2885 cm^{-1}$) was employed to indicate the presence of this

Fig. 3. Form stable phase change material aggregates (a) coated with (b) calcium hydroxide powder, (c) milk of lime, (d) pozzolana, and (e) cocciopesto.

polymer in the mortars, as it does not interfere with any other signal of the $CaCO_3$. From the intensity of this peak, it was concluded that, in the case of mortars containing the unmodified aggregates, PEG 1000 tends to migrate from the cross section to the surface of the mortar. On the other hand, the mortars containing the coated aggregates always exhibited a reduced intensity of the peak relative to PEG 1000, thus partly preventing the *washing-out* of this polymer. In particular, both calcium hydroxide and milk of lime coatings are able to effectively prevent the *washing-out* of PEG 1000 and, thus, its migration. Moreover, the aggregates coated with pozzolana and cocciopesto were able to partially confine PEG 1000, as its signal can be still observed in the FTIR spectra.

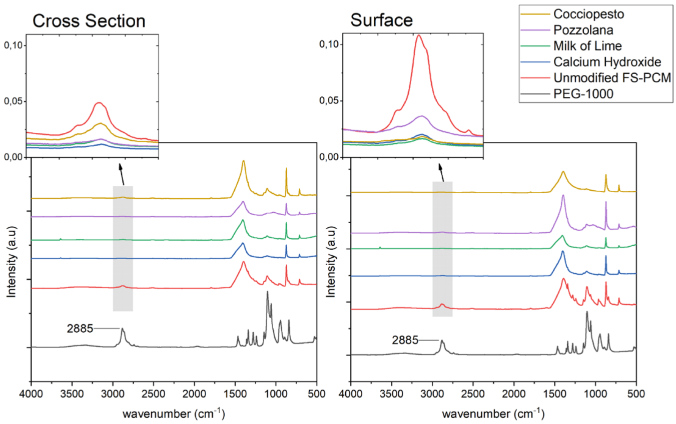


Fig. 4. FTIR-ATR measurements performed on a cross-section or surface of the mortars containing the unmodified FS-PCM or the four coated aggregates. In black, the reference spectrum of PEG 1000 is reported.

4 Conclusions

The presented study demonstrated that the impregnation method used to produce the PEG 1000-based FS-PCM was able to completely fill the porosity characteristic of the Lecce Stone inert support. On the other hand, PEG 1000 was found also on the surface of the PCM aggregates: this occurrence is probably responsible for the reduction of the adhesion between these aggregates and the binder composing the mortar. FTIR analysis revealed that the PEG 1000 composing the PCM is likely to migrate from the bulk to the surface of the mortar due to its high solubility in water.

In order to limit this phenomenon, thus improve the mechanical properties of the mortars containing a FS-PCM, different modifications were carried out on the PEG-based FS-PCM. Among others, the coating method proved to be an efficient and simple solution to confine the PEG 1000 inside the FS-PCM aggregates, avoiding its leaching. This method, in fact, was able to prevent the migration of the PEG 1000 present in the PCM to the surface of the mortar. This solution could also positively affect the thermal effectiveness of the PCM-based mortars, as more active material remains within the aggregates.

Further research is underway to assess the mechanical and thermal properties of mortars including modified FS-PCM aggregates, in order to confirm that this method is effective in limiting the reduction in mechanical properties of PCM-based mortars.

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