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Concrete-Polymer Composites in Circular Economy

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

Springer

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ISSN 2662-3161 ISSN 2662-317X (electronic)
Springer Proceedings in Materials
ISBN 978-3-031-72954-6 ISBN 978-3-031-72955-3 (eBook)
https://doi.org/10.1007/978-3-031-72955-3

This work was supported by Warsaw University of Technology and Building Research Institute.

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The registered company address is: Gewerbestrasse 11, 6330 Cham, Switzerland

Novel Sustainable Polymer-Based Phase Change Materials (PCMs) for Mortars Based on Different Binders for the Energy Efficiency of Buildings Located in Different Climatic Regions

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Abstract. A possible solution to reduce the consumption of fossil fuel and energy demand to power heating and cooling devices is represented by Phase Change Materials (PCMs). They can absorb, store and release energy according to their physical state that changes with the environmental temperature. In this work, novel eco-sustainable PCMs were developed through the form-stable method. Through this process, it was possible to create composite PCMs consisting of a natural inert matrix (i.e., a very porous stone obtained from processing waste) and an eco-friendly polymer, i.e., Poly-Ethylene Glycol (PEG). The composite PCMs were used to replace aggregates in mortars based on different binders (i.e., hydraulic lime, and cement). A complete characterization was performed on the new PCMs assessing their thermal stability and thermal efficiency. The study of the properties of the PCM-based mortars, in their fresh and hardened states, allowed to identify those with suitable mechanical properties. These latter were, then, subjected to a further investigation to assess their thermal behavior in response to different climatic loads. Encouraging results were achieved that allowed to establish the effectiveness of the novel PCMs in thermo-regulating an indoor environment.

Keywords: Phase Change Material (PCM) · Poly-Ethylene Glycol (PEG) · Mortars · Circular Economy (CE) · Energy efficiency

1 Introduction

To handle high energy demand, fossil fuel depletion and climate change, it is imperative to invest in the development of renewable energy systems as an alternative to conventional energy systems. Currently, energy storage systems are becoming increasingly well-known because their ability to improve the energy efficiency and energy savings was verified [1].

L. Czarnecki et al. (Eds.): ICPIC 2023, 61, pp. 594–602, 2025.
https://doi.org/10.1007/978-3-031-72955-3_61

In this area, Phase Change Materials (PCMs) play a key role as they can store excess energy by making it available when needed. Their integration into a building, through a suitable construction material (such as mortars) is one of the possible ways to improve the building energy efficiency while ensuring a good thermal comfort for the inhabitants [2]. Furthermore, a mortar occupies a large surface area in a building, and this allows a great storage capability [3].

Starting from these premises, two sustainable PCMs were developed with the aim to integrate them into plaster mortars. Low-cost, non-flammable, non-toxic polymers, i.e., Poly-Ethylene Glycol 1000 (PEG1000) and Poly-Ethylene Glycol 800 (PEG800) were selected as active PCM phases to be included in a porous matrix. The latter, a stone obtained in the form of small pieces from processing waste, was sieved to gain granules of an appropriate grain size. These granules were, then, vacuum-impregnated with the each PEG in liquid state, following the form-stable method [4]. Two different composite PCMs (i.e., LS/PEG800 and LS/PEG1000) were obtained, that were used as aggregates in mortars based on different binders (hydraulic lime and cement). The physical properties (i.e., workability and mechanical properties) of the developed mortars were evaluated. As expected, when one of the two form-stable PCM was included into a mortar, its mechanical performance decreased if compared to the same mortar without any PCM. Nevertheless, by adjusting the quantities of the mortar components, adequate mechanical characteristics were achieved. Further analyses were carried out on the optimized mortars to evaluate their thermal performance, in order to assess whether the presence of a PEG-based PCM can assure a proper thermal indoor comfort, saving energy and costs.

2 Materials and Methods

2.1 Materials

Two polymers with a different molecular weight were selected to produce the composite PCMs: Poly-Ethylene Glycol 1000 (PEG 1000), possessing a greater molecular weight and provided by Sigma-Aldrich Company (Germany) and Poly-Ethylene Glycol 800 (PEG 800) with a lower molecular weight and supplied by Wuhan Fortuna Chemical Co. (China). The different molecular weights of PEGs correspond to a different range of phase change temperatures: these two PEGs (i.e., PEG 1000 and PEG 800) were, in fact, selected on the basis of their phase change temperatures which should ensure the manufacture of two PCMs suitable for different climates. In addition, a third PCM was produced by combining the previous PCMs, to obtain a 50% mix, in order to extend the temperature range in which the melting/solidification process takes place. The thermal properties of the two single PCMs were measured through Differential Scanning Calorimetry (DSC), and the relative thermograms are shown in Fig. 1.

PEG 800 exhibits two endothermic peaks during the melting process: one pointed at around 18 °C and a smaller one at 25 °C and, at the same time, two exothermic peaks during the solidification process: one centered at around 13 °C and the other at 9 °C. The melting/solidification enthalpy is approximately of 150 J/g. The PCM based on this polymer was, then, selected for cold climates (i.e., continental temperatures). On the other hand, PEG 1000 shows an endothermic peak nearly at 43 °C and an exothermic peak close to 23 °C, with a latent heat in both heating and cooling stages of about 129 J/g.

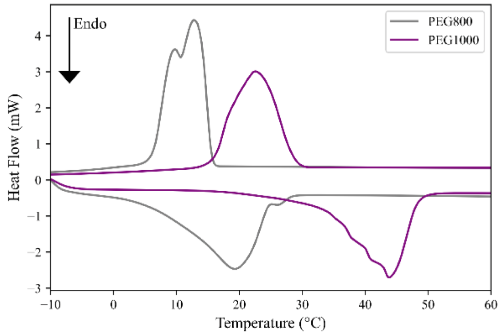


Fig. 1. Thermograms recorded through DSC on pure PEG 800 and pure PEG 1000.

The PCM manufactured with PEG 1000 was considered suitable for warm climates (i.e., Mediterranean/Tropical temperatures). The mix of the two PCMs, i.e. that based on PEG 800 and that produced starting from PEG 800, was expected to enlarge the range of phase change temperatures in order to be suitable for a wide variety of climate zones.

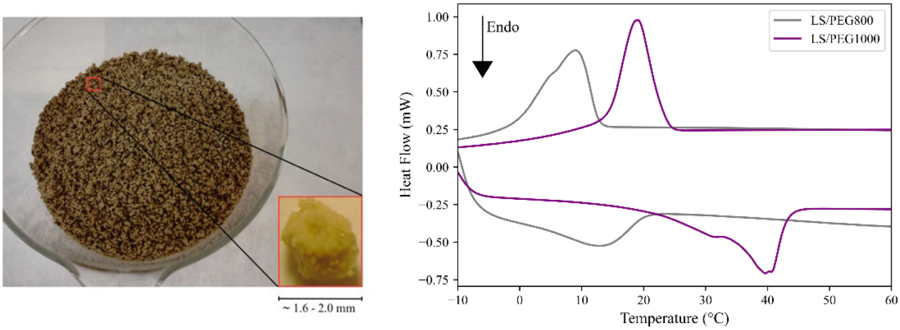
Lecce Stone (LS) was employed as matrix to contain the polymeric phase of the PCMs, i.e., each PEG, since it has a highly porous structure [5]. LS is a limestone very common in southern Italy and in fact it was supplied at km-0 from a local quarry. The processing waste of this stone (in the form of small flakes) were used with the aim of recycling this material in a circular economy perspective. The pieces of stone were, then, sieved to obtain appropriate size granules, i.e. between 1.6 and 2.0 mm.

Different binders were used to manufacture the mortar compositions, i.e., aerial lime, hydraulic lime, gypsum, and cement, as reported in [6–8]. However, the focus was pointed out on hydraulic lime and cement-based PCM containing mortars since they exhibited the greatest mechanical properties. A natural hydraulic-lime (HL), with a density of 2700 kg/m^3, was supplied by Cimpor (Lisbon, Portugal); a cement (C, CEM I 42.5 R), with a density of 3030 kg/m^3, was purchased by SECIL (Lisbon, Portugal). A polyacrylate (MasterGlenium SKY 627), supplied by BASF, was used as superplasticizer (SP) to keep low the water content with the aim of improving the mechanical properties of the mortars.

2.2 Preparation of the Form-Stable PCMs and Their Inclusion in Mortars

Two composite PCMs were produced starting by the selected PEGs (i.e., PEG 800 and PEG 1000) using LS as support matrix through the so-called *form-stable* method, by vacuum impregnation of the stone, as described in [5, 9]. In Fig. 2a, a PEG-based PCM is shown. The final PCMs obtained, indicated as LS/PEG800 and LS/PEG1000, were used to replace inert fine aggregates (usually sand) in the mortar formulations.

The composite PCMs were, then, thermally characterized to confirm their capacity to serve as thermal energy storage/release materials in the form of aggregates. The thermal tests were also employed to measure their melting/solidification temperature ranges and the relative latent heats. The results of DSC test performed on the LS/PEG800 and LS/PEG1000 PCMs are shown in Fig. 2b. The melting peak temperature for LS/PEG800



(a) (b)

Fig. 2. (a) Composite PCMs used as aggregates for mortars. (b) Thermograms recorded through DSC on the PCMs LS/PEG800 and LS/PEG1000.

is about 13 °C and the crystallization peak temperature about 9 °C; for the LS/PEG1000 composite, the melting temperature peak is about 40 °C and the crystallization one about 19 °C. For both PCMs, the enthalpy measured in both heating and cooling stages is about 29 J/g. The results of these tests confirmed that both PEG-based PCMs possess an adequate thermal behavior. They were, then, incorporated into the mortars based on both binders. Reference mortar formulations were also manufactured using LS particles as aggregate. The compositions of the different mix designs are summarized in Table 1.

As it can be seen from Table 1, an increased binder content (from 800 kg/m^3 to 1000 kg/m^3) and a higher amount of the SP (from 15 kg/m^3 to 20 kg/m^3) were employed in the study in order to improve the mechanical properties of the final mortars. The water at saturation point was used to completely impregnate the pure LS granules, i.e., to prevent them to absorb the water required to the mortars.

2.3 Methods

According to the European standard EN 1015-3, the workability of all the developed mortars reported in Table 1 was evaluated to determine their flowability in their fresh state, using the flow-table test. According to European standard EN 1015-11, the mechanical characteristics of the developed mortars were studied (in both flexural and compressive mode) employing a Lloyd dynamometer equipment. The mechanical tests were performed at a speed of 6 µm/s in flexural configuration and at a speed of 12 µm/s in compressive one, respectively. The results obtained on three specimens (at least) for each mortar were, finally, averaged.

3 Results and Discussion

The results of the tests performed on all the produced mortars (i.e., workability and mechanical characterizations) are presented in Table 2. In the same Table, the classification of each mortar, according to the standard NP EN 998-1, is also reported.

Table 1. Mortar formulations (indicated as kg/m^3)

Sample	Binder Content	Aggregates			SP	Water Saturation	Water	Water/Binder
		LS	LS/PEG800	LS/PEG1000				
HL_{800}_LS	800	1092	0	0	15	275	320	0.40
HL_{800}_LS/PEG1000	800	1729	0	398	15	0	375	0.47
HL_{1000}_LS	1000	682	0	0	20	171	380	0.38
HL_{1000}_LS/PEG800	1000	1082	249	0	20	0	320	0.32
HL_{1000}_LS/PEG800_LS/PEG1000	1000	1082	124	124	20	0	320	0.32
C_{800}_LS	800	1070	0	0	15	269	296	0.37
C_{800}_LS/PEG1000	800	1347	0	310	15	0	360	0.45
C_{1000}_LS	1000	772	0	0	20	194	390	0.39
C_{1000}_LS/PEG800	1000	1307	301	0	20	0	300	0.30
C_{1000}_LS/PEG800_LS/PEG1000	1000	1307	150	150	20	0	300	0.30

Table 2. Physical properties of the mortar formulations in fresh state (to determine the workability) and hardened state (to assess the mechanical properties).

Sample	Workability (mm)	Flexural Strength (MPa)	Compressive Strength (MPa)	Mortar Classification
HL_{800}_LS	165 ± 2.0	2.8 ± 0.5	17.0 ± 0.2	CSIV
HL_{800}_LS/PEG1000	175 ± 2.0	0.4 ± 0.1	1.5 ± 0.1	CSI – CSII
HL_{1000}_LS	175 ± 1.0	5.2 ± 1.5	11.7 ± 0.5	CSIV
HL_{1000}_LS/PEG800	170 ± 3.0	2.3 ± 0.4	3.5 ± 0.2	CSII – CSIII
HL_{1000}_LS/PEG800_LS/PEG1000	163 ± 2.0	2.1 ± 0.5	3.6 ± 0.5	CSII – CSIII
C_{800}_LS	160 ± 1.0	9.2 ± 0.9	26.3 ± 0.4	CSIV
C_{800}_LS/PEG1000	178 ± 3.0	1.9 ± 0.3	3.4 ± 0.8	CSII
C_{1000}_LS	180 ± 0.5	11.8 ± 1.1	65.6 ± 6.1	CSIV
C_{1000}_LS/PEG800	170 ± 1.0	2.1 ± 0.1	3.9 ± 1.2	CSII – CSIII
C_{1000}_LS/PEG800_LS/PEG1000	170 ± 4.0	2.0 ± 0.2	4.4 ± 0.7	CSII – CSIII

According to the results shown in Table 2, all the mortars presented excellent workability values, i.e. falling within the established range (160-180mm). Analyzing the results of the mechanical tests, as expected, the presence of a PCM caused reductions in the mechanical properties of mortars [10], irrespective to the type of PCM included in that mortar. However, it was possible to achieve suitable values of compressive strength when the both the binder (from 800 kg/m^3 to 1000 kg/m^3) and the superplasticizer (from 15 kg/m^3 to 20 kg/m^3) contents were increased. The developed mortars were, in fact, classified as CSII, CSIII, and CSIV, according to the EN 998-1 standard. These values resulted appropriate for the intended applications, i.e., the development of plaster mortars. The mortars were also subjected to DSC analysis to assess the presence of any PCM and its thermal characteristics. All the mortars containing a PEG-based PCM were found to be suitable from a thermal point of view for the intended climatic zone, in terms of melting/crystallization range of temperatures, as reported in [11]. The investigation moved, then, to the analysis of the thermal behavior of the mortars when subjected to a controlled temperature program. An experimental setup able to study the thermal behavior of the mortars applied on the wall of a small box in the four seasons of the year was realized, as illustrated in Fig. 3. The simulation of the four seasons took place in a climatic chamber, selecting two climatic conditions: those characteristics of Mediterranean regions and those typical of Continental area. Each mortar composition, reported in Table 1, was applied (thickness of 1cm) on the inner walls of a scaled test cell (20x20x20cm^3) composed by an insulating material (i.e., expanded polystyrene, XPS). A thermocouple (Type K) was placed in the center of the cell to measure the internal temperature during the experiment.

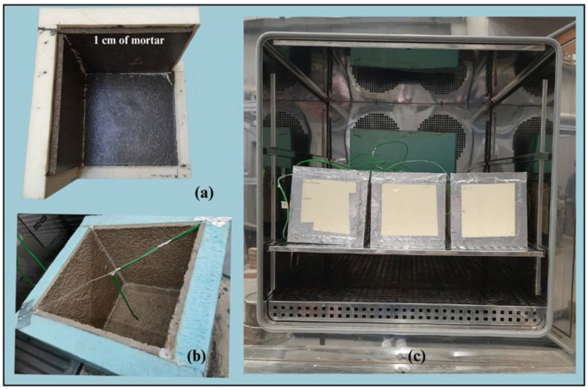


Fig. 3. Experimental setup employed to evaluate thermal performances: (a) section of a small-scale box with 1 cm-thick of mortar; (b) thermocouple centered inside the small-scale box; (c) climatic chamber where the small-scale boxes were placed to conduct the test.

The results obtained in the experiments performed on some of the PCM-based mortars, i.e., those containing PEG 1000, have been reported in [12]: they confirmed that the presence of the LS/PEG1000 composite PCM is able to positively influence the thermal behavior of the mortars. It was observed, in fact, that the maximum/minimum peaks were reduced/increased (especially in warmer seasons), with a consequent decrease in the energy needed for the indoor cooling/heating. The same tests have been recently carried out on mortars containing LS/PEG800 or LS/PEG800_LS/PEG1000 PCMS: the relative results has been just published in [13].

4 Conclusions

When a PCM is selected, it is extremely important to know its characteristic phase change temperatures (i.e. melting/crystallization) and latent heat capacity. In this work, two polymers with different molecular weights were selected to produce two PCMs: PEG1000 and PEG800. These PEGs were chosen for their sustainability characteristics and for their melting and crystallization range if temperatures considered appropriate for the intended applications, i.e. to produce indoor plaster mortars suitable for climatic conditions typical of Mediterranean regions and Continental areas. The polymers were included in a porous inert matrix (i.e., LS) through a vacuum impregnation process, obtaining as a result two composite PCMs, i.e. LS/PEG800 and LS/PEG1000. The PCMs were, then, used as aggregates in different mortars, i.e. based on hydraulic lime or cement. The addition of the PEG-based PCMs in mortars affected their physical properties in both fresh and solid states. However, with a suitable selection of the mortar composition, it was possible to achieve an appropriate workability and adequate mechanical properties. On the mortars that achieved the best mechanical performance the thermal performance was evaluated in order to assess their attitude to reduce the need for cooling and heating, from a building energy efficiency perspective.

Acknowledgements. This research was funded by PON Ricerca e Innovazione 2014–2020 Risorse React Eu - DM 1062/2021 Azione IV.4, "Dottorati e contratti di ricerca su tematiche dell'innovazione" and Azione IV.6, "Contratti di ricerca su tematiche green".

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