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Thermal transfer performance of a spherical encapsulated PEG 6000-based composite for thermal energy storage

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HIGHLIGHTS

- PEG 6000-epoxy composite is a solid–liquid PCM candidate for TES in buildings.
- PCM solidification is experimentally and modeling evaluated in a spherical module.
- The approach proposed to solidify the semicrystalline PEG is a two-solid front model.
- Analytical results agree reasonable with experimental data for PEG solidification.
- Guidance for PCM solidification in multi-spherical TES systems is provided.

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ABSTRACT

A polymeric phase change composite material (70 wt% polyethylene glycol, PEG, 6000)-epoxy resin (29 wt%) with aluminum nanopowder (1 wt%) as filler, P60-E, was developed and thermally tested first in a spherical macro capsule in order to be used in thermal energy storage (TES) systems in constructions with low energy consumption. Since the thermal behavior of the phase change component, PEG 6000, is highly influenced by its crystallization behavior, structural and thermal data were correlated. Consequently a high crystallinity degree of 82.6%, found by X-ray diffraction (XRD), for the PEG 6000 component is analogous with values obtained from integrated Raman spectra and DSC data (latent heat of -113.6 J/g) collected at a cooling rate of 0.4 °C/min. Both experimental and mathematical modeling of PEG 6000 solidification in the P60-E nanocomposite was conducted using a single spherical test cell. The heat transfer during solidification assumes time evolution of both liquid and the two solid radial fronts corresponding to crystalline chains of PEG and amorphous counterpart of PEG and epoxy resin in the P60-E composite. Good agreement between experimental values and calculated theoretical curves was found by using a two-front solids model.

1. Introduction

The building sector accounts for a significantly large contribution in the energy consumption (40%) and CO_2 emissions (36%) for developed countries across Europe [1]. TES systems for passive and active heating/cooling in buildings and use of recovered energies could help to reduce fossil fuel consumption and CO_2 emissions [2] while the thermal comfort of the residents is maintained [3]. The main advantages of the latent heat TES, LHTES, of the phase change materials, PCMs, comprise of its high energy storage density and ability to provide heat at a constant temperature [4–6].

Difficulties arising from applying organic PCM-containing systems are their low thermal conductivity, subcooling, change in density, poor thermal stability against extended cycling, corrosiveness, high price, phase separation, unpleasant odour, leakage during phase transition, etc. [6–8]. To improve low thermal conductivity of PCMs, carbon-type materials [9] and/or metallic conductors as aluminum, cooper and nickel fillers [10,11] are added to the PCM-containing systems.

In the last decades [6] various geometry PCM encapsulations have attracted significantly attention in order to try to mitigate a few of the above weaknesses. Apart from the increased area of heat transfer, a spherical encapsulated PCM, i.e. nano- [12], micro- [13] and macro-encapsulated PCM [3,14,15], is an effective way to enhance thermal conductivity, prevent direct contact of PCM with the environment which can cause corrosion [6] as well as leakage during solid-liquid phase transition. The most known technique for micro-encapsulation of

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Nomenclature

C_p	specific heat at constant pressure (J/g K)
f	fraction of latent heat of solidification
h_w	heat transfer coefficient (W/m^2 K)
m	mass (g, kg)
r	radius (m)
t	time (s, min)
Bi	Biot number
Fo	Fourier number
H_{ls}	latent heat of solidification (J/g)
H_{sl}	latent heat of melting (J/g)
H_{loss}	heat loss percentage (%)
Mu	Mu number
R	dimensionless radius $R = r/r_w$
Ste	Stefan number
T	temperature (°C, K)
Φ	constant of the model
X_c	crystallinity degree

Greek symbols

α	thermal diffusivity (m^2/s)
β	dimensionless rate
ε	relative deviation of theoretical vs experimental values (%)
φ	cooling rate (°C/min)
λ	thermal conductivity (W/m K)
ρ	density (kg/m^3)
Θ	dimensionless complex constant at interface

Subscripts

0	Initial
I	first stage of solidification
II	second stage of solidification
a	air
av	average value
l	liquid phase
m	mushy zone
s	solid phase
sph	sphere
w	external thermal exchange surface
S1	first solidification (crystalline), at liquidus front
S2	second solidification (amorphous), at solidus front
‘	first part of second stage
‘’	second part of second stage

organic PCMs [16], is the sol-gel method which is not suitable for water soluble PCMs. In this case macro-encapsulation of water soluble PCMs as polyethylene glycols (PEGs) [6,17] is required for thermal energy storage purposes.

A PCM has to meet some thermophysical, kinetic, chemical and economic criteria for a certain application [3–6,18]. Analogous, selection of capsule material is subject to few requirements as shell mechanical strength to prevent rupture, management of the PCM volume expansion, thermal stability, high thermal conductivity to enhance heat transfer, optical issue, inexpensive raw material and manufacturing processes [6,15]. One application of the packed bed latent heat storage systems with PCM-containing spheres [19] consists in energy efficient buildings [20]. It is well known that optimum design configuration lowers capital cost per kWht for a certain application [21]. Also, specific investment cost for a compact LHTES is predicted to be below 50 €/kWh until 2020 while its industrial application represents an objective for the next decade [4]. The dimensioning of TES systems with various geometries and efficiency estimation involve accurate knowledge of the thermal answer of the TES systems during charging and discharging processes by taking into account the climate parameters and necessary heat for the living space [3,8,20,22,23].

Gaps between experimental works and intricate transient thermal behavior of charging and discharging processes in the TES systems with organic PCM packed beds [3,19,21] are the main reasons for studying spherical encapsulated PCMs as PEGs. Also, expensive set ups of the packed bends [19,21] are other drawbacks to overcome. Our prior work [10] to obtain and thermophysically characterize three shape-stabilized PEG-epoxy resin-aluminum nanocomposites (PEG with molecular weights of 1000, 1500 and 2000) showed absorbed and released latent heat at about 100 J/g and at a discharge temperature bellow 55 °C. The larger molecular weight PEG was used, the higher latent heat was released [10,24]. Except for increased latent heat and melting temperature, the high molecular weight PEGs might show subcooling effects [10,17] which are detrimental for TES. Moreover, water solubility of the PEG component has to be prevented. To overcome this drawback, solidification of PEG 1500 component in the PEG 1500-epoxy-aluminum composite, P15-E, was tested in a laboratory test cell around a horizontal pipe with water as heat transfer fluid [25]. Its transferred heat was characterized by time evolution of the melt and two-solid fronts corresponding to crystalline and amorphous solids. To our knowledge study of spherical macro-encapsulated PEG 6000-epoxy composites in air for building purposes has not yet been reported in literature.

In this work, a mathematical model for solidification (discharging) of a new phase change nanocomposite material, PEG 6000-epoxy resin-aluminum, in a spherical polyethylene maro-capsule was developed, tested and validated against experimental data for further uses in packed beds for building heating. Influence of the crystallinity degree of the PEG 6000 component, obtained by DSC, XRD and in-situ Raman measurements, on thermal performances during discharging was discussed.

2. Experimental

2.1. Materials

P.a. reagents: polyethylene glycol of molecular weight of Mw = 6000 (Aldrich), epoxy resin Ropoxid 501 (Policolor, Bucharest) with three-ethylene-tetramine, TETA, (Policolor, Bucharest) as hardener and micronized Al powder (Riedel-de Haen) were melted together and mold transferred to yield the PEG 6000-Epoxy composite, named P60-E, as described earlier [10]. The weight percents of the components, PEG: Ropoxid 501:Al: TETA account for 70:26:1:3.

The P60-E mixture was transferred (Resin Transfer Molding) in a polyethylene sphere designed for experimental evaluation of a spherical TES system where a three-dimensional solid material polymerized inside during a 24 h span, the reaction being complete after 7 days. Description of the spherical module is presented further in Section 2.4.

2.2. Structure of the P60-E composite

Morphology and structure of the P60-E composite were investigated by scanning electron microscopy (SEM), X-ray diffraction and temperature-dependant Raman spectroscopy. Morphology of the P60-E composite was characterized by SEM FEI Quanta3DFEG model in high vacuum mode. Minimal sample preparation consisted in immobilization of the composite on a double-sided carbon tape. X-ray diffractograms were collected by means of a Rigaku diffractometer type Ultima IV in