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The manufacture of organic carbonate-poly(methyl ethylacrylate) nanowebs with thermal buffering effect

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ABSTRACT

In this study, novel nanowebs provided with thermal buffering properties were developed via coaxial electrospinning. Environmental friendly and nontoxic dialkyl and dialkenyl carbonates (DACs), namely, didecyl carbonate, didodecyl carbonate, ditetradecyl carbonate, dihexadecyl carbonate, dioctadecyl carbonate and dioleyl carbonate were synthesized from their fatty alcohols based on carbonate interchange reaction. Then the DAC-PMEA nanowebs were produced using 10–50 w% DAC in 3:1 chloroform-ethanol solution as core and 15 w% poly(methyl ethylacrylate) (PMEA) in ethanol as shell by coaxial electrospinning. The SEM images of the nanowebs presented randomly oriented, ultrafine fibers identified with smooth surfaces, continuous and cylindrical shapes. The nanowebs were able to provide considerable enthalpies (40–100 J g⁻¹) during their repeated phase changes, up to hundred heating-cooling cycles, corresponding to 52–92% efficiencies obtained from electrospinning. These shape stabilized, lightweight DAC-PMEA nanowebs showed a significant potential for the dynamic thermal insulation applications across various industries, especially for the protection of temperature sensitive materials.

1. Introduction

Phase change materials (PCMs) which absorb and release a great amount of heat repeatedly during phase conversions between solid–solid or solid-liquid phases have been receiving great attention in the last decade particularly to develop efficient thermal energy storage systems. Paraffin waxes, poly(ethylene glycols), fatty acids and their derivatives are the PCMs that have been widely studied in different fields such as textiles, buildings, electronics, solar energy systems, vehicle engines etc. [1]. Recently, organic carbonates (OCs), also known as dialkyl and dialkenyl carbonates (DACs), which have been widely used as nontoxic and noncorrosive replacements for hazardous chemical reagents in pharmaceuticals, cosmetics, electrochemical and catalytic reactions, have attracted attention as potential bio-based and renewable PCM candidates in consequence of their attractive thermal characteristics [2–9].

Temperature sensitive materials for instance fresh and cooked foods, medications and biotechnological products, quickly lose their potency if they are exposed to heating while transporting [10,11]. The temperature control of the commercial transport boxes is mostly dominated by the systems using ice, gel packs or dry ice as a coolant wherein the containers are insulated using expanded polystyrene (EPS).

However, their uses are limited due to the lack of small, lightweight, continuous, repeatable and cheap cooling mechanisms [11]. For conserving product quality and safety, the development of thermally controlled packaging (TCP) for transporting the temperature sensitive materials has gained great importance for both industry and researchers. TCP can be designed to play an active role to keep the temperature of the products within the acceptable temperature range during the time they spend in transit between the supplier's controlled environments to the customer's controlled environment, so as to meet the requirements of product safety, customer friendliness and cost-effectiveness. PCMs have been under investigation recently for TCP applications, because of their buffering effect against temperature changes in certain time intervals [12–18].

For most of the thermal management applications, PCM use in composites necessitates a prior shape stabilization of PCM in polymer or inorganic matrices for reducing its reactivity with the outside environment, preventing the leakage, decreasing the evaporation and diffusion rates and promoting the ease of handling [19,20]. A novel technique of coaxial electrospinning, currently developed for simultaneous electrospinning of two different solutions into core-shell structured nanofibres, has presented remarkable potential for use in PCM encapsulation applications. The selection of the shell polymer suitable

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for coaxial electrospinning is one of the most important tasks of the process, since the electrospinnability of the shell solution is responsible for the successful encapsulation of a core material regardless of core's electrospinnability under certain operating conditions. The viscosity and electrical conductivity of the shell solution must be higher than core's, and the surface tension between core-shell solutions must be low, over and above the effects of the operating parameters, such as flowing rates of core and shell solutions and the voltage applied between the injector and the collector [21].

Recently, several research groups have focused on the fabrication of PCM-polymer core-shell nanowebs using diverse types of PCMs as core material and a number of polymers as shell material by means of coaxial electrospinning in order to develop shape stabilized PCMs which can be easily applicable to various composites. McCann and his co-workers [21] reported the preparation of phase change fibers consisting of n-alkane cores, TiO₂-poly(vinyl pyrrolidone) (PVP) sheaths by melt coaxial electrospinning. The fibers consisting of 31 wt% hexadecane, 45 wt% octadecane and 36 wt% eicosane cores exhibited the enthalpy of melting of 71 J g⁻¹ at 16 °C, 114 J g⁻¹ at 27 °C and 88 J g⁻¹ at 37 °C, respectively. Chen et al. synthesized a series of diacid dioctadecyl esters (DADOEs) as novel solid-liquid PCMs, and then they produced bicomponent fibers of DADOEs and polyethylene terephthalate (PET) [22]. They observed that the morphology and thermal properties of composite fibers changed dramatically by adjusting the DADOE content and the type of DADOE, and measured the enthalpy of melting for DADOE/PET fibers between 11 and 67 J g⁻¹ at 45–48 °C. Chen et al. later produced ultrafine fibers of the polyethylene glycol/cellulose acetate (PEG/CA) composite, in which PEG10000 acted as the PCM and CA acted as the matrix, via coaxial electrospinning. The composite fibers displayed the latent heat of fusion of 86 J g⁻¹ at 58 °C and the latent heat of crystallization of 65 J g⁻¹ at 39 °C, respectively [23]. Do et al. produced PEG-poly(vinylidene fluoride) (PVDF) core-shell nanofibers via melt electrospinning using PEG1000/PEG2000/PEG4000 as the core material. The e-spun PEG-PVDF nanofibers loaded with 42.5 w% of PEG4000 and fabricated had the highest latent heat of melting of 68 J g⁻¹ at 63 °C among all [24]. Hu and Yu encapsulated natural soy waxes into polyurethane (PU) nanofibers using coaxial electrospinning technique. As a function of the wax concentration, the enthalpy of melting varied between 10 and 37 J g⁻¹ at a broad temperature interval of 20–57 °C [25,26]. Pérez-Masiá et al. encapsulated dodecane inside a food-derived protein of zein by coaxial electrospinning. They concluded that as-spun fibers displaying the enthalpy of fusion of 10–59 J g⁻¹ at about –10 °C, were quite suitable for food packaging applications [27]. Wen et al. prepared core-shell microfibers using commercial paraffin wax (RT27) as a core and poly(vinyl butyral) (PVB) as a shell material. The maximum melting and crystallization enthalpies of microfibers were about 128 J g⁻¹ at 28–31 °C and 124 J g⁻¹ at 25–24 °C respectively, and the corresponding encapsulation efficiency was as high as 70% [28]. Chalco-Sandoval et al. prepared electrospun coatings where they selected polycaprolactone (PCL) and polystyrene (PS) as encapsulating matrices of the commercial paraffin wax (RT5) for their application in refrigerated foods [29,30].

Golestaneh reported the fabrication of co-electrospun composite fibers of binary fatty acid eutectics of capric-lauric acid (CA-LA) and capric-palmitic acid (CA-PA) cores in PET shells. The fibers offered a wide phase-transition temperature range between –9 and 34 °C by absorbing about 56 J g⁻¹ heat during melting [31]. Very recently, Babapoor et al. fabricated coaxial electrospun PCM nanofibers using polyethylene glycol (PEG1000) as the core and polyamide 6 (PA6) as the shell material. They studied the effects of core flow rate and PEG content on the morphology, structure, and phase change behavior of the produced nanowebs [32].

Poly(methyl ethylacrylate) (PMEA) is a worldwide well-known copolymer for its excellent thermoplastic properties, low glass transition temperature, high thermal stability, protection from moisture, low gas permeability, resistance to acidic environment (pH < 5.5), easy dissolution in common solvents, non-toxicity, and cost effectiveness [33,34]. It has good filament-forming properties and electrospinnability, which raise its prospective application in industrial electrospinning processes [35].

The aim of this study is to develop novel nanowebs provided with thermal buffering properties using environmental friendly and non-toxic DAC cores and PMEA shells via coaxial electrospinning, suitable for controlled temperature packaging applications. The use of DACs (or organic carbonates) as PCM is quite new and there still exists a knowledge gap for their use in thermal management applications. PMEA is also promising to produce lightweight and shape stable PCMs in the form of nano or submicron scale webs. In addition, the manufacture of DAC-PMEA core/shell nanofibers, which has not been addressed yet, offer the advantages of renewable thermal energy and thermal control, reduced temperature fluctuations, ease of use and direct integration opportunity in packed products, all needed for the applications which concern with the environment and product safety. For this purpose, we first synthesized DACs and then produced nanowebs of DAC-PMEA bicomponent fibers for the best-suited production parameters of coaxial electrospinning. Structural and thermal characterizations of both organic carbonates and their electrospun nanowebs were then carried out throughout the comprehensive Fourier Transform Infrared (FTIR) spectroscopy, Nuclear Magnetic Resonance (NMR) spectroscopy, Scanning Electron Microscopy (SEM), Thermogravimetry-Differential Thermogravimetry (TG-DTG) and Differential Scanning Calorimetry (DSC) analyses.

2. Experimental

2.1. Materials

The reagents used for the synthesis of DACs, given in Table 1, were all technical grade (98%) and used without further purification (Sigma-Aldrich Inc.).

For the encapsulation of DACs via coaxial electrospinning, the copolymer of methacrylic acid and ethyl acrylate (PMEA) with the methacrylic acid groups of 46.0–50.6%, glass transition temperature of 96 °C ± 5 °C and the molecular weight of 320,000 g mole⁻¹ was

Table 1
Chemical reagents used in the syntheses of didecyl, didodecyl, dihexadecyl, dioctadecyl, and dioleil carbonates.

Chemical reagent	Formula	Molecular weight MW (g mole ⁻¹)	Function in the DAC synthesis
1-Decanol (C ₁₀ -capryl alcohol)	CH ₃ (CH ₂) ₉ OH	158.28	Saturated fatty alcohol, reactant
1-Dodecanol (C ₁₂ -lauryl alcohol)	CH ₃ (CH ₂) ₁₁ OH	186.33	Saturated fatty alcohol, reactant
1-Tetradecanol (C ₁₄ -myristyl alcohol)	CH ₃ (CH ₂) ₁₃ OH	214.39	Saturated fatty alcohol, reactant
1-Hexadecanol (C ₁₆ -palmityl alcohol)	CH ₃ (CH ₂) ₁₅ OH	242.44	Saturated fatty alcohol, reactant
1-Octadecanol (C ₁₈ -stearyl alcohol)	CH ₃ (CH ₂) ₁₇ OH	270.49	Saturated fatty alcohol, reactant
cis-9-Octadecen-1-ol (C ₁₈ -oleyl alcohol)	CH ₃ (CH ₂) ₇ CH=CH(CH ₂) ₇ CH ₂ OH	268.48	Unsaturated fatty alcohol, reactant
Diethyl carbonate	CH ₃ CH ₂ OCOOCH ₂ CH ₃	118.13	Dialkyl carbonate, reactant
Dibutyltin(IV) oxide	CH ₃ (CH ₂) ₃ SnO(CH ₂) ₃ CH ₃	248.94	Catalyst

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