



# Evaluation of paraffin infiltrated in various porous silica matrices as shape-stabilized phase change materials for thermal energy storage

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## ABSTRACT

Phase change behaviors of organic phase change materials (PCMs) confined in porous silica matrices, which are determined by the pore characteristics as well as interfacial interactions between PCM molecules and supporting solid, are crucial to PCMs for thermal energy storage performance. In this paper, a series of shape-stabilized phase change materials (ss-PCMs) were engineered with three silica matrices as support and paraffin as PCMs through solution impregnation. These porous silica matrices serve as an ideal skeleton for shape stabilization of melted paraffin PCMs and yield desirable thermal properties of paraffin confined in silica pores. The textural and chemical properties, crystallization, interfacial interactions, and thermal properties and stability are investigated using various techniques including nitrogen adsorption-desorption isotherms, Fourier transformation infrared (FT-IR) spectroscopy, small- and wide-angle X-ray diffraction (XRD), and scanning electron microscopy, as well as differential scanning calorimetry (DSC) analysis. The effect of the pore structure on crystallization of PCMs was studied and a novel phase change was unveiled. The phenomena of melting point depression and confinement effects are in line with theoretical thermodynamics, and the physical state of paraffin at the interfacial region in mesopore channel is analyzed considering pore geometric factors, revealing a non-melting interface layer in MCM-41. This evaluation of various silica matrices may provide important and general implications for the fundamental understanding of porous silica ss-PCMs performance with cost-effective and readily available raw materials.

## 1. Introduction

U.S. Energy Information Administration (EIA) predicts a 28% increase in world energy consumption between 2015 and 2040 in its International Energy Outlook 2017 [1]. Rising to this challenge is a daunting dilemma since it has also to be done in an environmentally friendly approach, involving an accelerated shift towards a highly or completely carbon-neutral society to obtain the 2 °C aim of the Paris Agreement [2]. To reduce carbon footprint, therefore, fossil fuels have to be repealed and replaced by renewable ones, which will play a fundamental role in addressing the ever-growing energy and environment problems [3]. Dramatic cost declines in solar and wind technologies [4], and now energy storage, open the door to the profound promotion of clean energy transformation [5,6]. Solar energy is the most abundant and promising renewable resource and is expected to contribute strongly to the future world energy mix. Much hope is pinned on solar energy utilization as a game-changer in attempts

towards low carbon energy industry. However, like other renewable ones, solar energy is intermittent and unstable. The conundrum of mismatch between solar energy demand and supply is one of the obstacles hindering its wide application.

Energy storage is a key aspect of dealing with the intermittent nature of renewable energy technologies and could be central to low-carbon energy systems [7]. However, stationary batteries are still perceived as unattractive for investors due to its cost [8]. So thermal energy storage (TES) using phase change materials (PCMs) have been a key area of research in the last three decades and more. PCMs exhibit a high heat of fusion as latent heat during melting and solidification with little volume change, which can be released later or even in different places. The idea of using PCMs became an important aspect of energy storage and management. Lower cost PCMs have emerged as a promising, cost-effective and technically robust approach to enhance the efficiency of energy storage for a variety of domestic and industrial sectors ranging from solar energy utilization [9], building energy

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Nomenclature		SM	silica matrix including MCM, EP and LJ
Abbreviations		$\Delta T$	temperature difference (K)
PCMs	phase change materials	$T_{\text{bulk}}$	bulk melting temperature (K)
ss-PCMs	shape-stabilized phase change materials	$T_r$	melting point (K)
XRD	X-ray diffraction	$\gamma_{\text{sl}}$	surface tension ( $\text{N m}^{-1}$ )
FTIR	Fourier transform infrared spectroscopy	$\rho$	particle density ( $\text{kg m}^{-3}$ )
TGA	thermogravimetric analysis	$r$	particle radius (m)
DSC	differential scanning calorimetry	$\Delta H$	phase change enthalpy ( $\text{kJ kg}^{-1}$ )
MCM	MCM-41	<i>Subscripts</i>	
EP	diatomite from EP Minerals, USA	x	paraffin:SM weight ratio
LJ	diatomite from Linjiang, China	m	melting
P	paraffin	f	freezing

conservation [10], textile [11] to automobiles industry [12].

The use of PCMs for thermal energy storage increases the power system flexibility by smoothing the mismatch between energy supply and demand through peak load shifting. During the development of PCMs, intense efforts have been directed at studying the relationship between the basic structure and phase change characteristics of many different groups of materials, which are majorly categorized as inorganic compounds, organic composites like paraffin, fatty acids, and some polymeric materials. PCMs has been critically examined to enhance performance and safety with respect to their premium energy storage [7,13–15]. Organic PCMs in general and paraffin wax in particular are a significant class of PCMs by virtue of unique thermal properties such as the ability to melt congruently, large working temperature range, good thermal stability and no obvious corrosion, thus making it the most suitable material for thermal regulation of building [16]. As with most energy technologies, PCMs for thermal energy storage faced challenges. Firstly, the ideal PCMs should meet a number of criteria with respect to thermophysical properties such as high enthalpy per unit volume, suitable phase change temperature, and no supercooling as well as long-term chemical stability and high thermal conductivity. Practical PCMs, for example, require their phase change temperatures to be within the temperature range for a given application and should possess high heat of fusion for practical energy storage. Moreover, the PCMs should be cheap enough to be readily available in large quantities. Actually, those merits are not fully possessed by most PCMs, and these shortcomings may degrade the system performance used for thermal energy storage and thermal regulation and can eventually hinder their final applications. However, advance in the synthesis and performance enhancement of novel composite materials in terms of nanomaterials [17], microencapsulation technology [18], porous solids packaging [19–21] as shape-stabilized PCMs (ss-PCMs) has opened new possibilities for PCMs.

To address the above problems involving actual utilization of PCMs, intense efforts have been devoted to developing packaging technology such as encapsulation or shape stabilization, which can preserve the

solid shape even at the temperature above the melting point of PCMs itself. This approach has been envisioned as a valid way to resolve the corrosion and leakage issues of organic PCMs [16]. It is worth to note that infiltrating organic PCMs into various porous matrices as ss-PCMs is a cost-effective alternative to the more expensive and time-consuming micro-encapsulation technology [22]. Porous matrices can yield an edge in ss-PCMs where scale and cost are critical, which would make it cheap and easy to scale up. Nowadays, a number of ss-PCMs have been reported by impregnating PCMs into various porous structures, including bentonite [23], zeolites [24], diatomite [20,25], carbon nanotube [26], expanded graphite [27], mesoporous silica [28–30]. Among which, porous silica matrices with a high specific area, large pore volume, excellent adsorption capacity, and chemical inertness, have attracted considerable attention in various fields including catalysis, environmental remediation, energy storage and conversion, and many multidisciplinary fields [31,32]. Table 1 summarized thermal properties of different porous silica materials as supporting matrices for ss-PCMs.

Extensive studies were conducted on porous silica matrices as the porous structure by a number of researchers and were found to be excellent candidates as supporting skeleton for ss-PCMs [25,33,34,38,39]. Porous silica ss-PCMs have unique features due to outstanding physicochemical properties stemming from their periodically arranged uniform pores in the sub-50 nm regime. The synthesis of ss-PCMs containing fatty acids and two hexagonal ordered porous silica (MCM-41, SBA-15) with nano-pores were presented and the nano-confinement effect was analyzed [30]. Due to the confinement effect, the phase-change enthalpy and phase-change temperature of the PCMs confined in a pore are different from those in bulk [27,37,39,40]. With respect to the advancement of ss-PCMs via impregnated into a porous carrier, critical factors like the porous network's major characteristics, such as pore size as well as pore shape, the geometric arrangement of pores must be evaluated, and chemical compatibility, availability in low cost are also important to consider. However, these reported contributions focus only on the influence of pore diameter. In fact, other aspects of

**Table 1**  
Summary of ss-PCMs based on porous silica materials in the literature.

Supporting matrices	PCM	Melting process		Solidifying process		Ref.
		$H_M$ ( $\text{Jg}^{-1}$ )	$T_M$ ( $^{\circ}\text{C}$ )	$H_S$ ( $\text{Jg}^{-1}$ )	$T_S$ ( $^{\circ}\text{C}$ )	
Silica nanosheets	Polyethylene glycol (PEG)	113.93	57.99	103.81	44.34	[33]
$\text{SiO}_2$ microspheres	Stearic acid	135.3	71.5	133.5	62.4	[34]
Silica gel	Hydrated salts	67.52	28.45	–	–	[35]
Diatomite	PEG	111.3	59.45	102.4	41.02	[25]
Diatomite	Paraffin	89.54	33.04	89.80	52.43	[36]
SBA-15	Stearic acid	69.8	69.1	68.1	66.7	[37]
SBA-15	Lauric acid	73.7	41.7	62.6	41.1	[30]
MCM-41		34.9	40.1	30.2	39.1	

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