

## SUPPORTING INFORMATION

Versatility of polyethylene glycol (PEG) in designing solid-solid phase change materials  
(PCMs) for thermal management and their application to innovative technologies

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The thermal properties of the organic form-stable PCMs reported in literature have been listed in Table S1. The heat storage efficiency ( $\gamma$ ) has been calculated in terms of heat loss percentage by using Eq. 1:<sup>1</sup>

$$\gamma = \left(1 - \frac{H_s}{H_m}\right) \times 100 \%$$

where  $H_s$  and  $H_m$  are solidification and melting enthalpy respectively.

With increasing percentage of heat loss, the efficiency of thermal energy storage decreases. The poly(acrylic acid)-PEG composite shows the lowest heat loss making it a highly efficient PEG based SSPCM whereas the composite of polyurethane prepared with PEG 6000 and graphene has high heat loss making it least efficient among the various organic form-stable materials explored. The use of porous supporting materials allows incorporation of high wt% of PEG thereby yielding high TES capacity.

Table S1 Thermal properties of organic form-stable PEG/PEO based PCMs.

Type of PCM	Hard segment	Soft segment	Enthalpy of fusion (J.g <sup>-1</sup> )	Phase change temperature (°C)	PEG wt%	Heat storage efficiency (%)	Reference
Cellulose-PEG	Cellulose	PEG 4000	142.9	52.2	40.0	12.5	2
CDA-PEG	CDA	PEG 4000	104.5	52.0	80.0	-	3
CDA-PEG	CDA	PEG 4000	155.0	52.0	85.0	-	4
Cellulose-PEO	Cellulose	PEO 13,060	134.7	63.4	25.0	5.4	5
Carboxymethyl cellulose- PEO	Carboxymethyl cellulose	PEO 13,060	140.2	58.4	25.0	1.5	5
Cellulose ether- PEO	Cellulose ether	PEO 13,060	156.8	64.7	25.0	2.2	5
Cellulose-PEG	Cellulose	PEG 4000	84.6	58.5	60.0	6.7	6

Agarose-PEG	Agarose	PEG 4000	110.8	57.7	70.0	10.6	6
Chitosan-PEG	Chitosan	PEG 4000	152.1	57.1	80.0	9.0	6
Cellulose-PEG-GNPs	Cellulose	PEG 6000	156.1	63.0	89.2	4.6	7
Cellulose acetate-PEG	Cellulose acetate	PEG 8000	155.3	60.5	96.5	-	8
PMMA-PEG	PMMA	PEG	141.6	56.9	80.0	1.1	9
Eudragit S-PEG	Eudragit S	PEG	144.2	50.9	80.0	1.8	9
Eudragit E-PEG	Eudragit E	PEG	148.8	56.9	80.0	2.6	9
PMMA-PEG	PMMA	PEG 20,000	121.2	58.0	90.0	10.6	10
Poly(acrylic acid)-PEG	Poly(acrylic acid)	PEG 10,000	135.1	56.3	90.0	0.7	11
Poly(ethylene-co-acrylic acid)-PEG	Poly(ethylene-co-acrylic acid)	PEG 6000	168.3	57.2	75.0	1.8	11
PMMA-PEG-AIN	PMMA	PEG 2000	115.6	46.4	70.0	-	12
PMMA-PEG-GNP	PMMA	PEG 2000	124.4	44.6	69.3	13.7	13
PU-BDO-Graphene	PU + BDO	PEG 6000	160.1	59.4	50.0	14.6	14
PU-n-eicosane	PU + BDO	PEG 10,000 and n-eicosane	141.2	37.9 and 57.4	63.7	9.4	15
PEG-MDI-glucose	Glucose	PEG 8000	131.9	61.1	70.0	7.9	16
PEG-active carbon	Active Carbon	PEG 6000	90.2	62.5	70.0	5.6	17
PEG-EG	EG	PEG	156.0	62.0	90.0	-	18
PEG-AC	AC	PEG	88.0	61.0	70.0	-	18
PEG-CMK-5	CMK-5	PEG	148.0	63.0	90.0	-	18

PEG alkyl ether-porous carbon	Porous carbon	Brij58 (M <sub>n</sub> =1124)	-	45.2	70.0	-	19
PEG-porous carbon from MOF	Porous carbon derived from MOF	PEG 4000	162.0	60.0	92.5	-	20
PEG-porous carbon from potato	Porous carbon derived from potato	PEG 4000	91.8	56.6	50.0	10.8	21
PEO-potato starch	Gelatinized potato starch	PEO 13,060	96.9	57.6	75.0	2.3	22
Glucose -PEG	Glucose	PEG	153	-	90.0	-	23
Lactose-PEG	Lactose	PEG	156	-	90.0	-	23
Fructose-PEG	Fructose	PEG	152	-	90.0	-	23
Epoxy-PEG	Epoxy	PEG 4000	132.4	54.2	75.0	-	24
PEG-poly(acrylonitrile-co-itaconate)	Poly(acrylonitrile-co-itaconate)	PEG 4000	118.5	53.9	73.0	2.3	25

Table S2 lists the thermal properties of hybrid form-stable PEG base PCMs. Fire-retardant composite of PEG-silsesquioxane has been found to be a very efficient material with low heat loss and good thermal energy storage capability. Cement restricts the crystallization of PEG to a great extent thereby incurring high enthalpy loss for PEG-cement composite. The composites of PEG with inorganic hybrid materials offers promising thermal energy storage capacity with increasing molecular weight of PEG reaching a high fusion enthalpy of 162.9 J.g<sup>-1</sup> with PEG 10,000 composite with SiO<sub>2</sub>.

Table S2 Thermal properties of hybrid form-stable PEG based PCMs.

Type of PCM	Hard segment	Soft segment	Enthalpy of fusion (J.g <sup>-1</sup> )	Phase change temperature (°C)	PEG wt%	Heat storage efficiency (%)	Reference
PEG-SiO <sub>2</sub>	SiO <sub>2</sub>	PEG 10,000	162.9	61.6	85.0	-	26
PEG-SiO <sub>2</sub>	SiO <sub>2</sub>	PEG 6000	102.8	56.5	80.0	1.0	27
PEG-SiO <sub>2</sub>	SiO <sub>2</sub>	PEG 10,000	74.5	59.3	80.0	-	28
PEG-SiO <sub>2</sub>	SiO <sub>2</sub>	PEG 2000	133.9	57.4	80.0	-	29
PEG-SiO <sub>2</sub>	SiO <sub>2</sub>	PEG 2000	122.0	36.0	80.0	3.0	30
PEG-SiO <sub>2</sub>	SiO <sub>2</sub>	PEG 4000	151.8	58.0	79.3	6.6	31
PEG-mesoporous matrices	AC	PEG 1500	102.0	48.3	80.0	5.4	32
PEG-mesoporous matrices	SBA-15	PEG 1500	83.0	44.0	80.0	7.1	32
PEG-mesoporous matrices	MCM-41	PEG 1500	80.0	47.0	80.0	4.7	32
PEG alkyl ether-porous silica	MCM-41	Brij 58 (M <sub>n</sub> =1124)	81.7	36.7	60.0	4.5	33
PEG alkyl ether-porous silica	MCM-41	Brij 76 (M <sub>n</sub> =711)	49.7	33.1	60.0	27.7	33
PEG-radial mesoporous silica	Mesoporous silica	PEG 4000	129.6	57.2	80.0	8.7	1
PEG-mesoporous silica	NH <sub>2</sub> -SBA15-CH <sub>3</sub>	PEG 2000	88.2	52.0	70.0	6.8	34
Montmorillonite-	Na-MMT	PEG 1000	104.8	32.9	44.4	2.1	35

PEG							
PEG-polymer clay nanocomposite	Epoxy	PEG 1800	149.4	56.2	60.0	22.8	36
PEG-organosiloxane containing phosphamide	Silsesquioxane	PEG 2000	124.7	50.8	81.8	0.6	37
PEG-diatomite	Diatomite	PEG 1000	87.0	27.7	50.0	5.5	38
PEG-diatomite	Diatomite	PEG 4000	105.7	57.9	58.0	9.6	39
PEG-diatomite	Diatomite	PEG 4000	103.7	59.0	55.0	11.2	40
PEG-calcium silicate	Calcium silicate	PEG 4000	122.1	57.0	70.0	12.5	41
PEG-cement	Cement	PEG 1000	23.8	24.3	25.0	44.0	42
Bischofite-PEG	Bischofite	PEG 2000	108.8	98.8	5.0	-	43
PEG-C <sub>3</sub> N <sub>4</sub>	C <sub>3</sub> N <sub>4</sub>	PEG 6000	-	-	40.0	-	44
PEG-CNIC	CNIC	PEG 6000	45.8	43.8	60.0	6.7	44

The thermal energy storage properties and average fiber diameter of reported electrospun fibers have been presented in Table S3. Biodegradable PEG-PLA electrospun fibres exhibit good thermal energy storage efficiency incorporating up to 66.7 wt% PEG. The highest phase change enthalpy was reported for Eu-PEG fibres having a value of 141.8 J.g<sup>-1</sup> at 63.9 °C and the lowest melting enthalpy was observed for PVA-PEG fibres.

Table S3 Thermal energy storage properties of electrospun fibers.

Type of PCM	Hard segment	Soft segment	Enthalpy of fusion (J.g <sup>-1</sup> )	Phase change temperature (°C)	PEG wt%	Average fiber diameter (nm)	Heat storage efficiency (%)	Reference
CA-PEG	CA	PEG 10,000	86.0	58.4	85.0	1579	24.2	45
CA-PEG	CA	PEG 10,000	36.7	52.1	-	-	29.5	46
CA-PEG	CA	PEG 1500	39.4	44.2	15.9	384	18.6	47
Polypropylene-poly(N-hydroxymethyl acrylamide)-PEG	Polypropylene	PEG 2000	17.0	54.6	14.0	-	-	48
PEG-poly(dl-lactide)	PLA	PEG	74.7	58.7	66.7	1540	11.5	49
PVP-Eu-PEG	Polyvinyl pyrrolidone	PEG 10,000	141.8	63.9	17.0	989	-	50
Polyamide-6-PEG	Polyamide-6	PEG 4000	58.0	52.0	30.0	104	-	51
Polyamide-6-PEG	Polyamide-6	PEG 1000	100.0	67.0	33.3	59	-	52
PVA-PEG	PVA	PEG 2000	27.8	48.3	22.2	-	-	53
PEG monoester of maleic anhydride	P(AN-co-AM)	MAPEG 4000	66.3	50.7	20.0	448	14.6	54



Table S4 lists the thermal characteristics of inherently thermally conductive PEG based form-stable PCMs. It can be inferred that the introduction of GNPs to PEG-GO composite affords high thermal conductivity of  $1.72 \text{ W.m}^{-1}.\text{K}^{-1}$  with high thermal storage efficiency. The MWCNT-PEG composite offers high enthalpy efficiency and hybrid graphene aerogels permit high fusion enthalpy of  $185.6 \text{ J.g}^{-1}$  with good thermal conductivity of  $1.43 \text{ W.m}^{-1}.\text{K}^{-1}$ . Almost all other composites have comparable heat loss percentages.

Table S4 Thermal characteristics of PEG based PCMs having inherent thermal conductivity.

Type of PCM	Hard segment	Soft segment	Enthalpy of fusion ( $\text{J.g}^{-1}$ )	Phase change temperature ( $^{\circ}\text{C}$ )	PEG wt%	Enthalpy efficiency (%)	Thermal conductivity ( $\text{W.m}^{-1}.\text{K}^{-1}$ )	Reference
PEG-EG	EG	PEG 1000	161.2	61.4	90.0	8.8	1.32	55
PEG-GO	GO	PEG 6000	156.9	62.3	90.0	-	-	56
PEG-GO	GO	PEG 6000	142.8	52.8	96.0	5.4	-	57
PEG-GO	GO	PEG 4000	174.5	62.9	96.0	9.2	-	58
PEG-GO-GNP	GO	PEG 10,000	178.1	64.9	96.0	8.1	1.72	59
PEG-GO	GO	PEG 6000	-	45.2	80.0	-	-	60
PEG-SG	SG	PEG 4000	165.1	58.0	96.0	-	1.04	61
MWCNT-PEG	MWCNT-OH	PEG 10,000	147.0	62.9	90.0	2.5	-	62
Hybrid graphene aerogel	Graphene aerogel	PEG 10,000	185.6	64.8	97.7	4.2	1.43	63
PU-graphite foam (GF)	GF	PEG 8000	80.3	46.1	-	-	3.5	64
PU-pitch based graphite foam	PGF	PEG 6000	60.3	43.8	73.0	-	10.86	65

Table S5 summarizes the thermal properties of various PEG based PCMs to which additives have been incorporated for improvement of thermal conductivity. The addition of GNP has been found to be highly beneficial in gaining high thermal conductivity of  $2.33 \text{ W.m}^{-1}.\text{K}^{-1}$ . On addition of MWCNT to PEG-SiO<sub>2</sub> composite, high enthalpy of  $135 \text{ J.g}^{-1}$  was achieved without hampering thermal energy storage efficiency of the composite.

Table S5 Thermal properties of PEG based PCMs having additives for improvement of thermal conductivity.

Type of PCM	Hard segment	Soft segment	Thermal conductivity enhancer	Wt% of thermally conductive filler	Enthalpy of fusion ( $\text{J.g}^{-1}$ )	Phase change temperature ( $^{\circ}\text{C}$ )	Thermal conductivity ( $\text{W.m}^{-1}.\text{K}^{-1}$ )	Reference
PEG-graft-cellulose	Cellulose	PEG 8000	Ni nanoparticles	6.0	51.0	60.0	0.64	<sup>66</sup>
mPEG-g-cellulose	Cellulose	mPEG 5000	EG	10.0	87.6	58.9	0.80	<sup>67</sup>
PEG-SiO <sub>2</sub>	SiO <sub>2</sub>	PEG 1000	AlN	20.0	129.5	61.1	0.56	<sup>68</sup>
PEG-SiO <sub>2</sub>	SiO <sub>2</sub>	PEG 6000	Cu	2.1	110.2	56.6	0.41	<sup>69</sup>
PEG-SiO <sub>2</sub>	SiO <sub>2</sub>	PEG 6000	MWCNT	3.0	135.1	53.3	0.46	<sup>70</sup>
PEG-PMMA	PMMA	PEG 2000	AlN	30.0	79.2	46.0	0.38	<sup>12</sup>
PEG-PMMA	PMMA	PEG 2000	GNP	8.0	114.1	41.9	2.33	<sup>13</sup>
PEG-diatomite	Diatomite	PEG	Ag nanoparticles	7.2	111.3	59.4	0.82	<sup>71</sup>
PEG-expanded vermiculite	Expanded vermiculite	PEG 4000	Ag nanowire	19.3	99.1	59.9	0.68	<sup>72</sup>

Table S6 Thermal properties of chemically modified PEG based SSPCMs.

Type of PCM	Hard segment	Soft segment	Linkage	Enthalpy of fusion (J.g <sup>-1</sup> )	Phase change temperature (°C)	Heat storage efficiency (%)	Reference
PEG-MDI-BDO	MDI and BDO	PEG 10,000	Urethane	138.7	65.2	9.0	73
PEG-MDI-PE	MDI and PE	PEG 10,000	Urethane	152.9	58.6	-	74
PEG-MDI-PVA	MDI and PVA	PEG 4000	Urethane	72.8	61.1	-	75
PEG-IPDI-BDO	IPDI and BDO	PEG 3400	Urethane	99.0	46.9	18.1	76
PEG-diisocyanates	HMDI	PEG 6000	Urethane	176.0	59.9	0.6	77
PEG-MDI-multibenzene ring tetrahydroxy compound (THCD)	MDI and THCD	PEG 6000	Urethane	137.4	56.1	7.1	78
PEG-MDI-TABE	MDI and TABE	PEG 6000	Urethane	153.5	48.4	6.2	79
PEG-MDI-β-cyclodextrin	MDI and β-cyclodextrin	PEG 8000	Urethane	115.2	60.2	3.1	80
PEG-MDI-trihydroxy surfactants	MDI and Tween 80	PEG 6000	Urethane	127.7	47.8	1.9	81
PEG-MDI-hexahydroxy compounds	MDI and sorbitol	PEG 8000	Urethane	107.5	59.7	4.2	82
PEG-HMDI-castor oil	HMDI and castor oil	PEG 6000	Urethane	117.7	51.4	7.3	83
PEG-diethanolamine-IPDI-BDO	Comb structured PU	PEG 5000	Urethane	121.9	59.0	3.1	84
PEG-MDI-diethanolamine	Thermosetting PU	PEG 6000	Urethane	98.2	49.9	3.8	85
PEG-MDI-MDEA-1,3-propanesulphonate	MDI and MDEA	PEG 10,000	Urethane	152.3	60.8	1.7	86
PEG-IPDI-cotton	IPDI and cotton	PEG 8000	Urethane	56.0	62.0	28.5	66
PEG-MDI-HB polyester	MDI and HB polyester	PEG 6000	Urethane	138.2	67.0	4.2	87
PEG-TDI-HB polyester	TDI and HB polyester	PEG 6000	Urethane	124.8	66.6	2.7	88
PEG-MDI-HB polyester	MDI and HB polyester	PEG 6000	Urethane	138.2	67.0	4.2	89
PEG-MDI-HB polyester-PE	MDI, HB polyester and PE	PEG 6000	Urethane	125.0	59.0	3.5	90

PEG-CDA	TDI and CDA	PEG 4000	Urethane	73.6	42.0	-	3
PEG-CDA	TDI and CDA	PEG 10,000	Urethane	142.0	62.0	-	91
mPEG-cellulose	Cellulose	mPEG 2000	Ether	149.1	50.4	11.1	92
mPEG-cellulose	MDI and Cellulose	mPEG	Urethane	153.6	58.9	0.3	67
mPEG-TDI-styrene	TDI and Styrene	mPEG	Urethane	108.5	57.7	24.7	93
Poly(PEG methyl ether methacrylate)	-	PEG methyl ether methacrylate (M <sub>n</sub> =2080)	Ester	132.5	44.7	10.4	94
Poly(decaglycerol-co-ethylene glycol)	Poly(decaglycerol)	PEG 4000	Ester	163.5	51.1	13.5	95
Crosslinked PEGA	-	PEGA 4050	Ester	141.3	52.4	13.4	96
Poly(PEG octadecyl ether methacrylate)	MMA	PEG octadecyl ether	Ester	73.0	41.1	10.9	97
PEG-PET	PET	PEG 6000	Ester	26.8	52.2	26.4	98
Eu-PEG	Eu coordinated with phenanthroline and terephthalic acid	PEG 2000	Ester	96.6	72.1	13.89	99
PEG-PAM	PAM	PEG 10,000	Ether	159.4 kJ/mol	58.9	13.2	100
PEG-poly(glycidyl methacrylate)	Poly(glycidyl methacrylate)	PEG 10,000	Ester	73.2	55.9	4.6	101
PEG-Polystyrene	Polystyrene	PEG 6000	Ether	179.4	44.9	19.9	102
PEG-Polyacrylonitrile	Polyacrylonitrile	PEG 4000	Ester	74.1	51.4	11.8	103
PEG-melamine-formaldehyde	Melamine-formaldehyde	PEG 6000	Amine	109.4	57.7	5.1	104
PEG-poly(styrene-co-acrylonitrile)	Poly(styrene-co-acrylonitrile)	PEG 4000	Ester	66.8	23.0	2.0	105
PEG-PHBV	PHBV	PEG 2000	Ester	134.3	56.0	19.28	106
Crosslinked PEG-poly(hydroxyethyl methacrylate)	Poly(hydroxyethyl methacrylate)	PEG 8000	Ether	144.5	60.9	-	107

Table S7 Thermal properties of microencapsulated PEG based PCMs.

Type of polymerization	Core material	Shell material	Particle size (nm)	Phase change temperature (°C)	Enthalpy of fusion (J.g <sup>-1</sup> )	Enthalpy efficiency (%)	Reference
In-situ	PEG 6000	UF	141 nm	41.9	17.8	-	108
In-situ	n-octadecane and PEG 600	UF	296.7 μm	28.6	3.9	58.9	109
In-situ	Na <sub>2</sub> CO <sub>3</sub> , PEG 1000 and n-hexadecane	UF	-	17.7	44.6	4.7	109
Suspension	PEG 800	Polystyrene	0.13 μm	0	0	0	110
Suspension	PEG 1000	Polystyrene	0.14 μm	0	0	0	110

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