High Thermal Conductivity Phase Change Composite with Percolating Carbon Fiber Network

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20 **Abstract**

21 Latent heat storage (LHS) using phase change materials (PCM) is a promising
22 technology for the effective use of solar and industrial exhaust heat. However, the heat
23 transfer rate of an LHS system is severely limited by the low thermal conductivity of the
24 PCM. Therefore, this paper describes the development of a high thermal conductivity
25 phase change composite (PCC) with a percolating network of a high thermal
26 conductivity filler. The relationship between the effective thermal conductivity of the
27 PCC and the network structure of the filler was investigated. The PCC were prepared by
28 the conventional melt-dispersion (MD) method and a novel hot-press (HP) method.
29 Erythritol (melting point: 118 °C, thermal conductivity: 0.73 W m⁻¹ K⁻¹) was used as the
30 PCM, and carbon fiber (thermal conductivity: 900 W m⁻¹ K⁻¹ in the fiber direction) was
31 used as the high thermal conductivity filler. The effective thermal conductivity of the
32 PCC was measured by the laser flash method and the network structures were observed
33 by energy dispersive spectroscopy using a scanning electron microscope. As a result, we
34 observed that the percolating filler network in the PCC could be easily formed by the
35 HP method, presenting a higher thermal conductivity with less filler additive than the
36 PCC fabricated by the MD method. Additionally, we found that PCM raw materials
37 with a high packing ratio accelerated the formation of the percolating filler network.

38

39 **Keywords:** Phase change material, Network structures, Latent heat Storage, Thermal

40 Energy Storage, Thermal conductivity

41

42 **Nomenclature**

43	A	Factor that depends on shape and orientation of filler	(-)
44	B	Factor that represents relative conductivity of both PCM and filler	(-)
45	$C_{P,CF}$	Specific heat at constant pressure of CF	(J g ⁻¹ K ⁻¹)
46	$C_{P,PCC}$	Specific heat at constant pressure of PCC	(J g ⁻¹ K ⁻¹)
47	$C_{P,PCM}$	Specific heat at constant pressure of PCM	(J g ⁻¹ K ⁻¹)
48	$k_{cal,PCC}$	Calculated value of effective thermal conductivity of PCC	(W m ⁻¹ K ⁻¹)
49	k_{CF}	Thermal conductivity of CF	(W m ⁻¹ K ⁻¹)
50	$k_{ef,PCC}$	Effective thermal conductivity of PCC	(W m ⁻¹ K ⁻¹)
51	k_{PCM}	Thermal conductivity of PCM	(W m ⁻¹ K ⁻¹)
52	$V_{f,CF}$	Volume fraction of carbon fiber	(-)
53	α_{PCC}	Thermal diffusivity of PCC	(m ² s ⁻¹)
54	ρ_{PCC}	Density of PCC	(kg m ⁻³)
55	ρ_{PCM}	Density of PCM	(kg m ⁻³)
56	$\phi_{i,PCM}$	Initial packing ratio of the PCM particles	(-)
57	$\phi_{m,CF}$	Maximum packing fraction of CF	(-)
58	ψ	Parameter that depends on volume fractions of PCM and CF	(-)

59

60 **Abbreviations**

61 CF Carbon Fiber

62 HP Hot Press

63 LHS Latent Heat Storage

64 MD Melting Dispersion

65 PCM Phase Change Material

66 PCC Phase Change Composite

67 SEM Scanning Electron Microscopy

68 EDS Energy Dispersive Spectroscopy

69

70 **1. Introduction**

71 Recently, latent heat storage (LHS) technology has played an important role in
72 sustainable energy infrastructures. LHS is typically based on the storage or release of
73 latent heat during a solid-liquid phase transition of phase change materials (PCM). This
74 have many advantages, such as that the latent heat provides a higher heat capacity than
75 sensible heat storage, the PCM can be a constant heat source at the phase change
76 temperature, and the reversible phase change processes allow for repeated use. Because
77 of these advantages, the LHS system has shown the potential to be applied in many
78 fields [1], such as building materials [2], battery thermal management [3], solar heat
79 utilization including concentrated solar power [4], and industrial exhaust heat recovery
80 [5-7].

81 To accelerate the heat transfer rate, the exploration of PCM with high thermal
82 conductivities is required. This is because typical PCM, such as paraffin, sugar alcohol,
83 and molten salt, have low thermal conductivities in the $0.1\text{--}1\text{ W m}^{-1}\text{ K}^{-1}$ range, which
84 has led to low charging and discharging rates of the LHS system. Several studies have
85 focused on developing phase change composites (PCC) with high effective thermal
86 conductivities by mixing high thermal conductivity fillers, such as carbon materials
87 [8-14], metals [15], ceramics [16], and nanostructured materials [17], with PCM.

88 Another route to increase the thermal conductivity is to impregnate PCM into high
89 thermal conductivity porous materials [18-22]. Among them, the preparation of PCC
90 using a carbon material is a promising method for enhancing the effective thermal
91 conductivity of PCM. Carbon materials are the best fillers for enhancing the effective
92 thermal conductivity since only a small volume fraction is needed, and they have high
93 thermal conductivity and low density. To date, there have been many studies on the
94 preparation of PCC using various types of carbon materials such as carbon fibers [8, 9,
95 23, 24], carbon nanotubes [8, 11], graphite [13, 22, 23, 25-27], and graphene [14]. Each
96 study has presented a remarkable increase in the effective thermal conductivity of PCC
97 with a carbon material as filler.

98 Of the utmost importance, is to form a percolating network of the conductive
99 filler to obtain a composite with a high thermal conductivity [28]. Generally, the
100 effective thermal conductivity of composite materials using fillers is related to the
101 connection state of the fillers. When the filler connection is percolated, the effective
102 thermal conductivity is significantly increased and the amount of filler used in this
103 situation is called the percolating threshold value. Therefore, it is important to form an
104 effective filler network to achieve the percolating threshold with less filler content.
105 However, few studies have deeply investigated the structure of the filler network, and

106 the method used to effectively form the percolating network is still a challenging issue.

107 In light of the above review, this paper describes the preparation of high
108 thermal conductivity PCC using a percolating network of carbon fiber (CF) as a high
109 thermal conductivity filler. The PCC were prepared by two methods, including the
110 conventional melting dispersion (MD) method and a novel hot-press (HP) method. The
111 relationship between the effective thermal conductivity of the PCC and the network
112 structure of the high thermal conductivity filler were investigated.

113

114 **2. Experimental**

115 We prepared PCM composites by two methods, the melting-dispersion (MD)
116 and hot-press (HP) methods. The critical difference between the two methods is that the
117 former mixed fillers in liquid PCM, and the latter mixed fillers with solid PCM
118 particles.

119 **2.1 Materials**

120 Erythritol ($C_4H_8O_4$) was used as the PCM, as it has a phase transition
121 temperature T_m of 118°C and a thermal conductivity of $0.733 \text{ W m}^{-1} \text{ K}^{-1}$ [15]. Table 1
122 lists the thermophysical properties of this PCM.

123 Carbon fiber (CF, XN-100, Nippon Graphite Fiber Co., Ltd) was used as the
124 high thermal conductivity filler. Table 2 lists the specifications of the CF. The CF has a
125 diameter of 10 μm , length of 50 μm , and density of 2220 kg m^{-3} . The anisotropic
126 thermal conductivity in the fiber direction is $900 \text{ W m}^{-1} \text{ K}^{-1}$ and in the vertical direction
127 is $10 \text{ W m}^{-1} \text{ K}^{-1}$.

128 **2.2 Preparation of the PCC**

129 **2.2.1 Conventional Melting-dispersion Method**

130 Fig. 1 a) illustrates the procedure to prepare PCC by MD. First, the PCM was
131 heated and melted at 150 °C in a thermally insulated beaker on a hot plate. Different

Table 1: Thermophysical properties of erythritol as the PCM.

Material	Thermal conductivity [W m ⁻¹ K ⁻¹]	Melting point [°C]	Latent heat [J g ⁻¹]	Specific heat [J g ⁻¹ K ⁻¹]	Density [kg m ⁻³]
Erythritol	0.73	118	340	1.68	1450

Table 2: Specifications of carbon fiber as the high thermal conductivity filler.

Material	Thermal conductivity [W m ⁻¹ K ⁻¹]	Specific heat [J g ⁻¹ K ⁻¹]	Density [kg m ⁻³]	Diameter [μm]	Length [μm]
Carbon fiber (CF)	900	12	2220	50	12

132

133

Fig. 1 Schematic diagram for producing a PCC composite by a) the melting dispersion method and b) the hot-press method along with the sample preparation for laser flash measurements.

134 amounts of CF were dispersed into the liquid PCM once at 150 °C. Each mixture was
135 then agitated at 1500 rpm for 30 min with a stirrer. Next, the melted PCC were removed
136 using a dropper, and then, were poured into a cylindrical aluminum (Al) mold with a
137 diameter of 10 mm. Finally, the products were removed from the molds, cut, and
138 polished to form disks with diameter of 10 mm and height of 2 mm.

139 **2.2.2 Hot-press Method**

140 Fig. 1 b) shows the procedure to prepare the PCC by HP. First, the PCM
141 particles and CF were wet-mixed in 99.9% acetone at 1500 rpm with a stirrer, for 30
142 min at room temperature. Then, the well-mixed wet samples were dried. Next, about 3 g
143 of the dry sample was transferred to a carbon mold with a diameter of 12.7 mm and then
144 placed in a press machine with an electric heater. Fourth, the sample was hot-pressed at
145 130 °C at 10 kN for 1 h, and then, the hot-pressed sample with diameter of 12.7 mm and
146 height of about 10 mm was cooled and removed from the mold. Finally, a sample with a
147 size of 10×10×1 mm was cut from the horizontal direction to the press-direction and
148 polished carefully.

149 We used 5 types of PCM particles with different particle size distributions as
150 the raw materials for the preparation of the PCC by HP. We assumed that the particle
151 distribution would influence the configuration of the PCM and CF in the PCC since

solid, not liquid, PCM particles were mixed with the CF. Fig. 2 a) shows the particle distribution of the PCM particles. The difference in the particle distributions led to the difference in the initial packing ratios of the PCM particles $\phi_{i, PCM}$. Here, the $\phi_{i, PCM}$ was measured with JISA1104 method [29] based on Japan industrial standards committee . Fig. 2 b) illustrates the concept of the $\phi_{i, PCM}$. In this way, we also investigated the effects of the $\phi_{i, PCM}$ on the effective thermal conductivity of the PCC and the filler-network structure.

Fig. 2 a) Particle size distributions of the PCM particles used in HP and b) a schematic showing the concept of the packing ratio of PCM particles, $\phi_{i, PCM}$. Note that different particle distributions result in different $\phi_{i, PCM}$ s.

162 2.3 Characterization Techniques

163 The density of the PCC was measured by a pycnometer (QUANTACHROME
164 INSTRUMENTS, Ultrapycnometer 1000). The accuracy was ± 0.02 %. The
165 measurement was repeated for 5 times for each specimen, and the deviation was
166 controlled within ± 0.01 %. The average value of the measurement was applied. The
167 real volume fraction of CF, $V_{f,CF}$ in the final PCC was calculated based on the measured
168 density of the PCC. Note that the real amount of CF in the final PCC differed from the
169 initial amounts of CF weighed for preparing PCC. This is because of that a part of the
170 CF was attached on wall of a beaker during mixing, leading to the loss of CF. The real
171 volume fraction, $V_{f,CF}$, of CF in PCC was calculated using the following Eq. (1):

172

$$173 \quad \rho_{PCC} = (1 - V_{f,CF})\rho_{PCM} + V_{f,CF}\rho_{CF} \Leftrightarrow V_{f,CF} = \frac{\rho_{PCC} - \rho_{PCM}}{\rho_{CF} - \rho_{PCM}} \quad (1)$$

174

175 where ρ_{PCC} , ρ_{PCM} , and ρ_{CF} are the density of the PCC, PCM, and CF, respectively.

176 To evaluate the effective thermal conductivity of the PCC, the thermal
177 diffusivity of the PCC α_{PCC} was measured using the laser flash method thermal constant
178 measurement system (ULVAC, Inc. TC-7000) at room temperature. The accuracy of
179 thermal diffusivity measurement was ± 5.0 %. The measurement has been repeated for

180 10 times for each specimen (deviation: 5.5 %), and the average value was applied.

181 The effective thermal conductivity of the PCC $k_{ef,PCC}$ was evaluated based on the
182 following relationship [25]:

183

$$184 \quad k_{ef,PCC} = \alpha_{PCC} \cdot \rho_{PCC} \cdot C_{P,PCC} \quad (2)$$

185

186 where $C_{P,PCC}$ is the specific heat of the PCC. $C_{P,PCC}$ was estimated using the following
187 equation:

188

$$189 \quad C_{P,PCC} = \frac{V_{f,CF} \cdot \rho_{CF} \cdot C_{P,CF} + (1 - V_{f,CF}) \cdot \rho_{PCM} \cdot C_{P,PCM}}{V_{f,CF} \cdot \rho_{CF} + (1 - V_{f,CF}) \cdot \rho_{PCM}} \quad (3)$$

190

191 The laser flash method is broadly used to measure thermal diffusivity of solid materials
192 for the convenient operation. This method was applied to measure the thermal
193 diffusivity of the PCC in previous studies with good results [30, 31]. On the other hand,
194 some researchers have noted that the surface roughness of the specimen greatly
195 influences the measurement accuracy because the thermal contact resistance between
196 the specimen and sensor modifies the measurement [32] for transient methods, such as
197 the laser flash method. Therefore, in this study, the PCC specimens were carefully

198 polished with #2000 emery paper, and then, the carbon was uniformly coated to reduce
199 the thermal contact resistance.

200 To observe the filler network structure, the elemental distribution from the
201 cross section of the PCC was observed using energy dispersive spectroscopy (EDS)
202 under the view of a scanning electron microscope (SEM, JEOL, JSM-7400F).

203 **3. Results**

204 **3.1 Effective Thermal Conductivity of the PCC**

205 Fig. 3 shows the effective thermal conductivity of the PCC prepared by the MD
206 and HP methods, together with calculated values from Nielsen's equation [33, 34] (see
207 **Appendix A**). The effective thermal conductivity of the PCC prepared by both methods
208 increased with increases in the volume fraction of the CF. The effective thermal
209 conductivity of PCC by MD was well-matched to the calculated value. On the other
210 hand, the effective thermal conductivity of the PCC by HP increased exponentially and
211 was much higher than those fabricated by MD. In addition, the PCC prepared by HP
212 using the PCM particles with a higher packing ratio provided a higher effective thermal
213 conductivity.

214

215

Fig. 3 Effective thermal conductivities of the phase change composites prepared by the hot-press method and melt-mixing method, together with calculated values from Nielsen's equation [33, 34] (see **Appendix A**).

Fig. 4 shows the EDS mapping from the cross-section of the PCC prepared by MD with CF contents of a) 12 vol%, b) 17 vol%, and c) 22 vol%. Fig. 5 presents the EDS mapping from the cross-section of the PCC, which have a different volume fraction of CF, as prepared by HP method. For the HP samples, the PCM particles with different packing ratios were used as the raw materials. Sample a) has a lower packing ratio of 0.59, and sample b) has a higher packing ratio of 0.71. Erythritol is composed of carbon (C), hydrogen (H), and oxygen (O), while CF contains only carbon. Therefore, we can distinguish the dispersion condition of the CF in the PCC with the EDS maps, as indicated by the different color for C (red) and O (blue). Significant differences in the

226 CF configuration/structure were observed in the PCC prepared by MD and HP. In the
227 PCC prepared by MD, the CF was uniformly dispersed in all the PCC samples.
228 Although a high concentration of CF was observed in the sample with the highest
229 volume fraction of CF, percolating networks of CF were not observed even at 20 vol%
230 of CF. On the other hand, CF appeared to be allocated near the PCM particles of the
231 PCC fabricated by HP. The connected structure of the CF increased as the CF
232 concentration increased, and a percolation was observed in the PCC with 22 vol% of
233 CF.

234

235

Fig. 4 SEM-EDS images of the phase change composites with CF contents of 12, 17, and 22 vol% prepared by the melting dispersion method.

Fig. 5 EDS mapping from the cross-section of the PCCs, which have different volume fractions of carbon fiber $V_{f,CF}$ prepared by the hot-press method using PCM particles with a) a lower packing ratio ($\phi_{i,PCM}$) of 0.59 and b) a higher ϕ_{PCM} of 0.71 as the raw materials.

Fig. 6 shows the comparison of the EDS mapping of the PCC with similar volume fractions of CF (18-21 vol%), but different packing ratios of the PCM particles, focusing on the regions of high CF concentration compared to Fig. 5. Remarkable differences in the CF configuration between low and high packing ratios of the PCM particles were observed. In the PCC with a low packing ratio (0.59), it was observed that the CF was only allocated around large PCM particles with diameters of about 200-400 μm . In the PCC with a middle packing ratio (0.65), the medium PCM particles with diameters of about 100-200 μm were located between large PCM particles with diameters of about 200-400 μm , and the CF was allocated around them. In the PCC with a high packing ratio (0.71), the PCM particles with small to medium diameters of about 50-200 μm were located near the CF, between the larger PCM particles.

Fig. 6 Comparison of the EDS mapping of the PCCs with similar $V_{f, \text{CF}}$ (18-21 vol%) and different packing ratios of PCM $\phi_{i, \text{PCM}}$ focusing on high carbon concentrations compared to Fig. 5.

254 4. Discussion

255 There are two main reasons that the thermal conductivities of the PCC prepared
256 by HP were much higher than the PCC prepared by MD: 1) the difference in the
257 orientations of the CF and 2) the difference in the network structures of the CF in the
258 PCC.

259 In the PCC prepared by MD, the CF was randomly oriented, as shown in Fig. 4,
260 since it can move freely in the liquid PCM during MD fabrication. On the other hand,
261 there was a possibility that the CF was vertically oriented with respect to the direction
262 of the press for the PCC prepared by HP. This is because CF, which has a high aspect
263 ratio, tends to lie within a plane perpendicular to the pressing stress. This was also
264 observed in PCC with expanded graphite prepared by a cold compression method [12].
265 This orientation accelerated the drastic increase of the effective thermal conductivity of
266 the PCC since the CF has an anisotropic thermal conductivity. In this case, the thermal
267 conductivity in fiber direction is $900 \text{ W m}^{-1} \text{ K}^{-1}$, but that in the circumferential direction
268 is about $10 \text{ W m}^{-1} \text{ K}^{-1}$. In conclusion, when compared to MD, HP was determined to be
269 a more effective method to prepare the high thermal conductivity PCC using anisotropic,
270 high thermal conductivity fillers such as CF.

271 Comparing the CF configuration/structure of the PCC prepared by MD to those

272 prepared by HP, significant differences were observed. The percolating network of CF
273 was not observed in the PCC prepared by MD even with a 20 vol% of CF, while the
274 percolating network of CF was clearly observed in the PCC prepared by HP with a CF
275 content over 20 vol% (see Fig. 4 and Fig. 5). This was the main reason that the thermal
276 conductivity of the PCC prepared by MD only gradually increased with an increase of
277 the volume fraction of CF, but that those prepared by HP increased in an exponential
278 manner, as shown in Fig. 3. Many researchers, especially in the field of high thermal or
279 electrical conductivity composites for plastics, focus that the formation of a percolating
280 network of filler is a trigger of the drastic increase in effective thermal and electrical
281 conductivity of the composite materials [28, 35, 36]. In previous studies of high thermal
282 conductivity composites, the packing ratio and shape factor of the filler materials are
283 mainly investigated [34] since the composites were usually prepared by MD or similar
284 methods. Various models for estimating the thermal conductivity [34, 37-40] of the
285 composites have determined that the shape characteristics of the filler are important
286 parameters to enhance the thermal conductivity. However, for discussion of the PCC
287 prepared by HP, an additional shape parameter of the PCM matrix should be taken into
288 consideration. According to the results of this study, percolation of the CF filler was
289 easily achieved in the PCM matrix with a higher packing ratio. Furthermore, the

290 packing ratio of the PCM was governed by controlling the particle size distribution of
291 the raw PCM particles. As shown in Fig. 5, the CF was naturally located around the
292 PCM particles during HP. This was because the PCM particles were in solid form
293 during HP and unmovable. In other words, when the matrix material was solid when the
294 filler was mixed with it, the configuration of the filler was strongly influenced by the
295 shape characteristics of the matrix materials. Fig. 5 provides conclusive evidence of this
296 the assumption, and we can clearly observe that the CF was located near the PCM
297 particles. It was considered that the percolating network structure of the filler was easily
298 formed in this condition since, initially, the PCM particles were in contact with each
299 other, and the void of PCM particles filled by the CF was originally percolated. This is
300 similar to water percolating into a packed bed of spheres. From these discussions, we
301 can conclude that HP has a great advantage in that it can easily form percolating
302 structures of high thermal conductivity fillers. This result may agree with a study of
303 Zhu et al. [41] that points out non-uniform distribution of filler lead to a better thermal
304 performance in heat change and discharge processes.

305 It is an important and interesting result that the PCC with a higher $\phi_{i,PCM}$ had a
306 lower percolating threshold value and higher thermal conductivity, as shown in Fig. 3.
307 This is closely related to the difference of filler network structures between the PCC

Fig. 7 Simplified model of the relationship between the PCM particles as a matrix material and CF as filler: a) the PCC with a lower packing ratio and b) the PCC with a higher packing ratio of PCM particles.

308 with a higher and smaller $\phi_{i,PCM}$, as clearly observed in Fig. 6. Fig. 7 shows the

309 simplified model of the relationship between the PCM particles (orange) as the matrix

310 material and CF (black) as the filler, based the experimental observation as shown in Fig.

311 6. Fig. 7 a) demonstrates the PCC with a lower packing ratio of PCM and Fig. 7 b)

312 depicts the PCC with a higher packing ratio of PCM. Different from the lower packing

313 ratio, the PCC with a higher packing ratio has a special filler configuration indicating

314 that large voids were formed by the contact of the large particles and medium matrix

315 particles filled the voids. Moreover, the voids were percolated. As shown in Fig. 5 and 6,

316 the CF naturally filled in the voids; therefore, a smaller amount of CF can percolate in a

317 higher packing ratio. In this way, this study clarified that the matrix particles with a

318 higher packing ratio have the advantage of the formation of a percolating network of

319 filler. This new concept is unique compared to the conventional concept that focused

320 only on the shape and structure of the fillers.

321 This suggests that the thermal conductivities of the composites increase with a
322 smaller amount of filler with matrix particles with high packing ratios. Although the
323 highest value of packing ratio investigated in this study was 0.71, a preparation of
324 higher packing ratio are expected according to literature about the packing ratio of
325 particles [42-44]. Therefore, the formation of a percolating network with a smaller filler
326 volume was expected with this higher packing ratio. This new concept can potentially
327 be applied various composites because it is reasonable to control the packing ratio of
328 matrix particles, such as PCM and polymers, compared with the shape of the filler
329 materials, such as CF and graphene.

330

331 **4. Conclusions**

332 This paper describes the preparation of high thermal conductivity PCC with a
333 percolating network of CF. The PCC were prepared by a conventional MD and a novel
334 HP method. The relationship between the effective thermal conductivity of the PCC and
335 the network structure of the CF was investigated. The main results are provided below.

336

337 1. HP is a more effective method than MD for preparing high thermal conductivity PCC
338 using anisotropic, high thermal conductivity fillers, such as CF.

339

340 2. The CF tends to orient in a plane perpendicular to the pressing stress for the PCC
341 prepared by HP. This orientation accelerated the drastic increase in the effective thermal
342 conductivity of the PCC since CF has an anisotropic thermal conductivity.

343

344 3. Using PCM particles with a high packing ratio as the raw material, a percolating
345 network of CF was found with a smaller amount of filler in the PCC prepared by HP.

346 In conclusion, we observed that a percolating network of filler in a PCC can be
347 easily formed by HP, and the HP method can achieve PCC with high thermal
348 conductivity using a smaller volume fraction of the filler than the PCC fabricated by

349 conventional MD. These results indicate the feasibility of using a new PCC with a high
350 thermal conductivity and percolating network of CF, which will contribute to the
351 efficient use of solar energy and industrial waste heat.

352

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356

357 Appendix A

358 We used Nielsen's model, which is used when dealing with conductive
 359 composites formed from short fiber/particulate fillers, to predict the effective thermal
 360 conductivity of PCC in Fig. 3. This model considers not only the thermal conductivity
 361 and concentration of the continuous/dispersed phases, but also the orientation, packing
 362 ratio, and the aspect ratio of the filler [33, 34]. This model was used to predict the
 363 calculated value of effective thermal conductivity of PCC, $k_{cal,PCC}$:

364

$$365 \quad k_{cal,PCC} = k_{PCM} \left(\frac{1 + A \cdot B \cdot V_{f,CF}}{1 - B \cdot \psi \cdot V_{f,CF}} \right) \quad (A1)$$

366

367

$$368 \quad B = \frac{\frac{k_{CF}}{k_{PCM}} - 1}{\frac{k_{CF}}{k_{PCM}} + A} \quad (A2)$$

369

$$370 \quad \psi = 1 + \left(\frac{1 - \varphi_{m,CF}}{\varphi_{m,CF}^2} \right) V_{f,CF} \quad (A3)$$

371 Here, k_{PCM} and k_{CF} is thermal conductivity of PCM and CF, A is a factor that depends
 372 on the shape and orientation of the filler, B is a factor that represents the relative

conductivity of both the PCM and the filler, ψ is a parameter that depends on the volume fractions of both components, and $\varphi_{m,CF}$ is maximum packing fraction of the filler material. The values of the parameter A of 11 and $\varphi_{m,CF}$ of 0.52 were used in this study, according to the literature [33, 34]

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