Characterization and Application of Nanomaterials 2024, 7(1), 4912.
https://doi.org/10.24294/can.v7i1.4912

Article

Enhancing the thermal properties of paraffin wax as latent heat storage material using hybrid nanomaterials

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CITATION

El-Sebaii AA, Aboul-Enein S, Ramadan MRI, et al. Enhancing the thermal properties of paraffin wax as latent heat storage material using hybrid nanomaterials. Characterization and Application of Nanomaterials. 2024; 7(1): 4912. https://doi.org/10.24294/can.v7i1.4912

ARTICLE INFO

Received: 29 February 2024
Accepted: 25 March 2024
Available online: 18 April 2024

Abstract: Paraffin wax is the most common phase change material (PCM) that has been broadly studied, leading to a reliable optimal for thermal energy storage in solar energy applications. The main advantages of paraffin are its high latent heat of fusion, low melting point that appropriate solar thermal energy application. In addition to its accessibility, ease of use and ability to be stored at room temperature for extended periods of time. Nevertheless, improving its low thermal conductivity is still a big noticeable challenge in recently published work. In this work, the effect of adding nano-Cu_2O, nano-Al_2O_3 and hybrid nano-Cu_2O-Al_2O_3 (1:1) at different mass concentrations (1, 3, and 5 wt.%) on the thermal characteristics of paraffin wax is investigated. The measured results showed that the peak values of thermal conductivity and diffusivity are achieved at wight concentration of 3% when nano-Cu_2O, nano-Al_2O_3 are added to paraffin wax with significant superiority for nano-Cu_2O. While both of those thermal properties are negatively affected by increasing the concentration beyond this value. The results also showed the excellence of the proposed hybrid nanoparticles compared to nano-Cu_2O, and nano-Al_2O_3 as it achieves the highest values of thermal conductivity and diffusivity at weight concentration 5.0 wt.%.

Keywords: thermal characteristics; phase change materials; paraffin wax; hybrid nanocomposites

1. Introduction

Paraffin wax (PW) is one of the most important materials used as phase change materials (PCMs) in the thermal energy storage system [1]. A lower thermal conductivity is considered the main disadvantage of PCMs which causes lower heat transfer rate during the charging and discharging processes. Many studies have been conducted to overcome this property, such as adding metallic or nonmetallic nanoparticles with high thermal conductivity [2], inserting fins [3], fibrous materials [4], macro-micro and nano-encapsulations [5,6], metal foams [7], carbon nanotubes [8].

The above-mentioned techniques used to improve the thermal conductivity of the PW include the addition of high conductive nanomaterials which is the simplest and most practicable technique. The influence of adding nano-graphite (NG) on the PW thermal conductivity was experimentally studied by Li [9]. The results showed that the thermal conductivity of the composite PCMs with 1% and 10% NG is 2.89 times and 7.41 times that of pure paraffin. Nano-Silicon nitride (Si_3N_4) at different mass fractions (1, 2, 3, 4, 5, 10 wt%) was studied by Yang et al. [10] to enhance the thermal properties of PW. They observed an improvement of 35% in thermal conductivity and also found an improvement in thermal diffusivity reaching 47% at 10 wt% Si_3N_4

additional fraction. Wang et al. [11] experimentally studied the effect of dispersing copper oxide (CuO) as a nanomaterial at different mass fractions of 0.3, 0.6, 0.9 and 1.2% into the PW. They concluded that the thermal conductivity of nanocomposite PCMs with a weight fraction of 1.2% had improved by 24.4%. Pise et al. [12] experimentally evaluated the improvement of thermal performance of PW integrated with nano-alumina (Al_2O_3) particles at various mass concentrations of nanoparticles of 1, 3, and 5%. They reported that the thermal performance of paraffin wax is enhanced up to 14%, compared to pure paraffin. Another experimental study concerning the thermophysical properties of Al_2O_3 nanoparticles/paraffin emulsions with two mass fractions of 5 wt.% and 10 wt.% [13]. The measured results showed that the increase in thermal conductivity is nonlinear with the increase of nanoparticle mass fraction. Yanqi et al. [14] examined how interfacial thermal resistance and particle size affected the thermal conductivity of paraffin/expanded graphite (EG) composites. They found that the increase in thermal conductivity is directly correlated with larger particle sizes. The small EG particles have less of an enhancement in thermal conductivity because interfacial thermal resistance predominates in their impact on the composite thermal conductivity. Sari et al. [15] also examined the paraffin/EG composite material and revealed that the thermal conductivity of the composite increased to 0.82 W/mK at 10 wt% EG. Huang et al. [16] investigated the enhancement of thermal conductivity of paraffin composites using non-equilibrium molecular dynamics simulation. The simulation results indicated that the thermal conductivity of PW is significantly enhanced by adding graphene-oxide which is more efficient than graphene. Maher et al. [17] investigated the effect of the addition of nanosilicon carbide (SiC) and nanosilver (Ag) based Paraffin composites on the thermal characteristics of PCM. The results revealed that the thermal conductivity of the paraffin/SiC composite improved by 58.2% which is much higher than the thermal conductivity of the paraffin/Ag composite that improved by 31.2% at the same mass fraction of 15 wt%. Qusay et al. [18] found that the thermal conductivity of the PW is improved by 18.2% when adding 3 wt% of nano-SiC into the PW. The impact of silver nanoparticles on the thermal conductivity of the PW was experimentally studied by Pradeep et al. [19]. Their results showed that thermal conductivity increases with the increase of the mass concentration of Ag nanoparticles. Sahan et al. [20] concluded experimental research evaluating the effect of adding nanomagnetite (Fe_3O_4) on the thermal properties of PW. They found that the thermal conductivity increased by 48% and 67% when adding 10 wt% and 20 wt% nano magnetite, respectively.

According to several studies, PCM could benefit from the addition of two or more hybrid nanoparticles [21–23]. When hybrid nanoparticles are used instead of single nanoparticles, the researchers conclude that the thermal conductivity increases at the same additional mass fraction compared to individual nanoparticles. From this point forward, several practical, numerical, and experimental investigations focus on the use of hybrid nanoparticles. Kumar et al. [24] studied the influence of hybrid nanoparticles containing SiO_2 and CeO_2 nanoparticles on the thermo-physical characteristics of the PW as PCM with various mass fractions (0.5, 1.0, and 2.0 wt%). They observed that the highest paraffin's thermal conductivity (0.298 W/m K) is achieved at 2.0 wt%. Kalbande et al. [25] carried out the addition of CuO and multi-walled carbon nanotubes (MWCNT) hybrid nanoparticles into the PW for thermal energy storage applications. They found that the thermal conductivity of nano-enhanced paraffin wax (PCM) was 6.125% higher than that of pure paraffin wax. Harikrishnan et al. [26] investigated the effect of dispersed hybrid nanoparticles CuO–TiO_2 into PW which includes several mass concentrations of 0.25%, 0.5%, 0.75% and 1 wt% to improve its thermal performance. They reported that the optimum studied concentration of hybrid nano-phase change material (HnPCM) is 1.0 wt%, which the improvement of thermal conductivity reaches 46.81% compared to pure paraffin. Ibrahim et al. [27] added nano-TiO_2, nano-MgO, and a 50% mixture of the two kinds into PW at different mass fractions of 0.25%, 0.5%, 0.75% and 1 wt% to find the maximum thermal storage characteristics of PCM. They observed that the highest enhancement is achieved when adding 1% of the nanoparticles and the thermal conductivity at this fraction is 4.6%, 3.9%, and 4.6% for nano-TiO_2, nano-MgO and hybrid nanoparticles, respectively.

In this work, the experimental investigations were conducted thoroughly to analyze the variation of thermo-physical properties of paraffin wax based as PCM under the influence of various weight concentrations (1%, 3%, 5%) of the nanoparticles, namely: Cu_2O and Al_2O_3. It is very important to mention here that Cu_2O nanoparticle is used for the first time, as authors know, to improve the thermal properties of paraffin wax. A thorough investigation into the impact of additional hybrid nanoparticles on the thermo-physical properties of paraffin is also rare in the literature. Therefore, the hybrid nanoparticles were also prepared by mixing equal masses of Cu_2O and Al_2O_3 to study their effect on the thermo-physical characteristics of the PCM. It was found that the hybrid nanoparticles at a mass fraction of 5.0 wt.% presented significant potential for enhancing the thermal storage properties of the paraffin wax.

2. Experimental work

In this study, the effect of adding Cu_2O, Al_2O_3 nanoparticles and hybrid-Cu_2O-Al_2O_3 nanoparticles into PW under different mass fraction concentrations (1%, 3%, 5%) on its thermal characteristics has been studied. To determine the accurate weights of pure PW, Cu_2O, Al_2O_3, and hybrids of the two nanoparticles, a digital balance with an accuracy of 0.0001 g was employed. Using a water path, 100 g of pure PW was melted at a melting point of 56 ℃ in order to prepare a specific PW/nanocomposite to obtain paraffin/nano-Cu_2O (NPCM-1), paraffin/nano-Al_2O_3 (NPCM-2) and paraffin/hybrid nano-Cu_2O-Al_2O_3 (1:1) (NPCM-3) as shown in **Table 1**. After the completely melted PW, nano-Cu_2O, nano-Al_2O_3 and hybrid nanoparticles were added into PW individually under continuous stirring for around 15 min to reduce the precipitation of the droplet's nano-additives into the PW and make the mixture homogenous. These steps were repeated at each of the nano-additives with various mass concentrations (1%, 3%, 5%). All samples are allowed to cool at room temperature and then shaped into a disc shape in the press designed for this process, where all prepared samples of PW, NPCM-1, NPCM-2 and NPCM-3 were impressed at 15 mm diameter and 3 mm thickness as shown in **Figure 1**. The thermal properties of PW before and after adding the nano-additive materials were measured using the Hot Disc Transient (Hot Disc TPS 500 S). Repeating the test twice produces more accurate, very flexible, fast, non-destructive, and reliable thermal properties of PW before and after adding the nano-additive materials including thermal conductivity, thermal diffusivity, and specific heat. The size of the samples must be determined when a suitable disc radius for a particular material has been chosen and the best test times for the disc and material combination are known. Taking into consideration that the nickel spiral sensor 7577 radius of 2.1 mm with Kapton insulation sandwiched between two sample portions to ensure tight contact with the sensor to minimize the amount of air gap. More details about Al_2O_3 nanoparticles preparation and characterization can be found elsewhere [28]. While Cu_2O nanoparticles are purchased from Qualikems Fine Chem Pvt. Ltd, Vadodara, Gujarat, India.

Table 1. Sample labelling and compositions.

S. No.	**Sample label**	**Composition (wt.%)**
1	Pure paraffin	100 PW
2	1% NPCM-1	99 PW + 1.0 Nano-Cu_2O
3	3% NPCM-1	97 PW + 3.0 Nano-Cu_2O
4	5% NPCM-1	95 PW + 5.0 Nano-Cu_2O
5	1% NPCM-2	99 PW + 1.0 Nano-Al_2O_3
6	3% NPCM-2	97 PW + 3.0 Nano-Al_2O_3
7	5% NPCM-2	95 paraffin+ 5.0 Nano-Al_2O_3
8	1% NPCM-3	99 PW +0.5 Nano-Cu_2O + 5.0 Nano-Al_2O_3
9	3% NPCM-3	97 PW + 1.5 Nano-Cu_2O + 1.5 Nano-Al_2O_3
10	5% NPCM-3	95 PW + 2.5 Nano-Cu_2O + 2.5 Nano-Al_2O_3

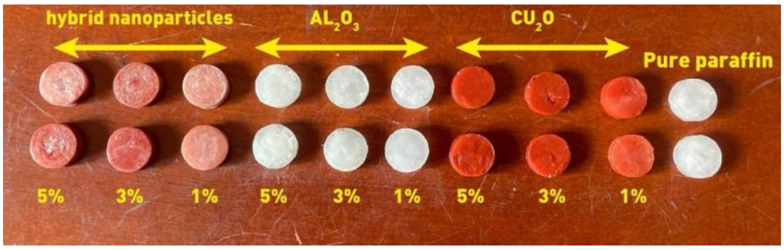

Figure 1. Photograph of the prepared paraffin nanocomposites.

3. Results and discussions

In this section, the measured values of different thermal properties of the prepared samples of pure paraffin and nanocomposites are presented, discussed and evaluated. The compromise between adding individual or hybrid nanoparticles into the PW is also of great interest in this section.

Figure 2 shows the variation of the thermal conductivity of NPCM-1, NPCM-2 and NPCM-3 with the concentrations. For samples NPCM-1 and NPCM-2, it was observed that the thermal conductivity of samples NPCM-1 and NPCM-2 reaches its peak value of 0.2760 W/m K and 0.2708 W/m K at 3.0% mass fraction of nano-Cu_2O and nano-Al_2O_3, respectively, with an improvement of 10.98% and 9.27% compared to the measured value for pure PW (0.2457 W/mK). But such a tendency is declined at 5.0 wt.% for both samples. The improvement of thermal conductivity is calculated as:

$$percentage\ of\ improvement = \frac{k_{nanocomposite} - k_{pure\ paraffin\ wax}}{k_{nanocomposite}} \times 100,$$

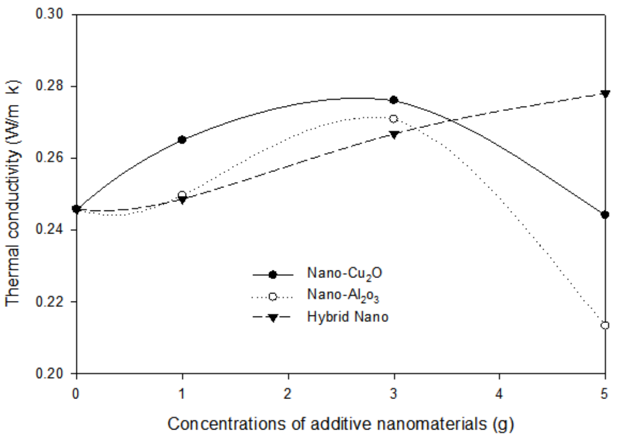

Figure 2. The thermal conductivity of the NEPCMs at different concentrations of 1.0, 3.0 and 5.0 wt.%.

The improvement of thermal conductivity of the PW is primarily because these additive nanomaterials have higher thermal conductivity and the combination performance of the nano-additives into the paraffin wax to enable the use of phase-change heat at higher temperatures to enhance the released rate of heat in paraffin. The results of **Figure 2** also shows that the thermal conductivity of PW/nano-Cu_2O is higher than that of PW/nano-Al_2O_3 at all studied mass concentrations. While it is higher than those values measured for PW/hybrid nanoparticles except at a mass fraction of 5.0%. This situation of decline happens at higher concentrations (>3%) in NPCM-1 and NPCM-2 samples due to the poor combination between the nano-additives and paraffin causes an interfacial thick layer [29] hence augmenting the interface thermal resistance between the paraffin and nano-additives. This layer is unable to contribute to the phase change temperature as phase change does not take place in the interface layer, so it decreases the thermal storage unit volume. Furthermore, the thicker thermal storage layer also diminishes the heat conduction performance of the NEPCMs. The same non-linear trend of thermal conductivity of nano Cu–paraffin composites has been reported by Lin and Al-Kayiem [30]. The results reveal that the increased mass percentage of nanoparticles in the base material may form their repressible agglomeration due to the prevailing cohesive forces, which would account for the diminished slope in the magnitude of thermal conductivity at the upper mass fractions. The phenomenon of an increase and sudden decrease in thermal conductivity of nano-Al_2O_3 was also reported by Arshad et al. [31] due to randomly molecules motion within the disordered microstructure of paraffin in the liquid phase.

The hybrid nano-Cu_2O and nano-Al_2O_3 show different effects as shown in **Figure 2**, where the thermal conductivity increases with incrusting the mass fraction (under the studied values) to reach 0.2780 W/m K at a mass concentration of 5.0% compared to 0.2441 W/mK, 0.2133 W/mK for nano-Cu_2O and nano-Al_2O_3, respectively. This improvement achieved by adding hybrid nanomaterials is considered the best among the studied cases which reaching 11.62% compared to pure paraffin. Keep in mind that this improvement was achieved at a higher concentration (5.0%). The results of **Figure 2** also show that the improvement percentages of the thermal conductivity of paraffin/nano-Cu_2O, paraffin/nano-Al_2O_3 and paraffin/hybrid nanoparticles at 1.0 wt.% are 7.28%, 1.56%, and 1.17%, respectively, compared to pure paraffin. While these improvements reached 10.98%, 9.27%, and 7.87%, respectively at a mass concentration of 3.0% compared to pure paraffin. The results also showed a drop of 0.65%, and 13.19% in the thermal conductivity of PW/nano-Cu_2O, and PW/nano-Al_2O_3 at 5.0 wt.%, respectively. **Figure 2** also shows that the thermal conductivity improvement for PW/nano-Cu_2O is higher by about 5.8%, and 1.88% than for PW/nano-Al_2O_3 and PW/hybrid nanoparticles, respectively at mass fraction 1% while these percentages increase to 6.19%, and 3.37% respectively when mass fraction 3.0 wt% is used. On the other hand, the thermal conductivity of PW/hybrid nanoparticles at 5 wt.% is higher than the corresponding measured values of PW/nano-Cu_2O, and PW/nano-Al_2O_3, which are 12.19% and 23.27%, respectively.

Among the physical properties, thermal diffusivity is considered one of the most important properties because it measures the facility of a material to conduct thermal energy corresponding to its ability to store thermal energy. Considering the fact that thermal diffusivity has a great significance in thermal management, this property was investigated in the current study as shown by the results presented in **Figure 3**. This figure shows that the thermal diffusivity of samples NPCM-1 and NPCM-2 reach their maximum values of 0.1595 m^2/s and 0.1252 m^2/s at 3.0% mass fraction of nano-Cu_2O and nano-Al_2O_3, respectively, compared to 0.1130 m^2/s for pure paraffin. So, the maximum improvements achieved by adding nano-Cu_2O and nano-Al_2O_3 are 29.15% and 9.74%, respectively, compared to pure paraffin. Accordingly, the thermal conductivity is directly proportional to the thermal diffusivity, so the behavior observed in thermal diffusivity is the same in thermal conductivity, increasing the mass fraction of nano additives causes an increase in the thermal diffusivity except at concentration of 5.0 wt.% for each of the nano-Cu_2O, nano-Al_2O_3 where the decline occurs, it decreasing to 0.1006, 0.0692 m^2/s, respectively, in comparison to pure paraffin.

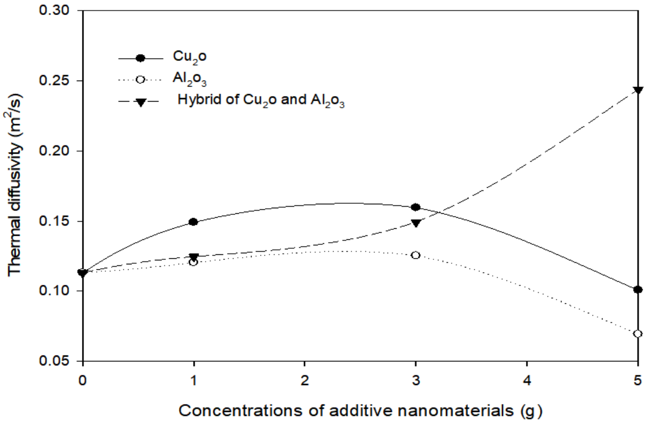

Figure 3. The thermal diffusivity of the NEPCMs at different concentrations of 1.0, 3.0 and 5.0 wt.%.

From the results of **Figure 3**, the thermal diffusivity enhanced to 0.1245 m^2/s, 0.1493 m^2/s and 0.2440 m^2/s at 1.0, 3.0 and 5.0 wt.%, respectively. In comparison to nano-Al_2O_3 and hybrid nano, the paraffin wax/nano-Cu_2O composite exhibits a percentage improvement of 19.21%, and 16.39% at weight concentration of 1%, and 21.50%, and 6.39% % at weight concentration of 3%, respectively, as shown in **Figure 3**. However, the hybrid nano has a percentage rise of 58.77% and 71.64% greater than nano-Cu_2O and nano-Al_2O_3 at a weight concentration of 5.0%, respectively. This means that the higher the dispersion of suspended nanoparticles causes higher thermal diffusivity which accelerated heat transmission from the top to the bottom of the nanocomposite. The limited dispersion of suspended nanoparticles will affect the value of thermal diffusivity, which will create a reduced heat transfer rate even though the ratio of added nanoparticles is higher. Comparison between thermophysical properties of all established samples is summarized in **Table 2**.

Table 2. Enhanced thermal properties of the pure paraffin wax.

Samples	**Thermal conductivity (W/mk)**	**Percentage of enhancement (%)**	**Thermal diffusivity(m^2/s)**	**Percentage of enhancement (%)**
Pure paraffin	0.2457	-	0.1130	-
1% NPCM-1	0.2650	7.28	0.1489	24.11
3% NPCM-1	0.2760	10.98	0.1595	29.15
5% NPCM-1	0.2441	-	0.1006	-
1% NPCM-2	0.2496	1.56	0.1203	6.07
3% NPCM-2	0.2708	9.27	0.1252	9.74
5% NPCM-2	0.2133	-	0.0692	-
1% NPCM-3	0.2486	1.17	0.1245	9.24
3% NPCM-3	0.2667	7.87	0.1493	24.31
5% NPCM-3	0.2780	11.62	0.2440	53.69

The specific heat is the only measured property that decreased with the nano-Cu_2O, nano-Al_2O_3 and hybrid additions into PW as it is inversely proportional to the thermal conductivity and diffusivity as shown in the following equation:

$$\alpha = \frac{k}{\rho c_p} m^2/_{s}.$$

where α and k are thermal diffusivity (m^2/s), and thermal conductivity (W/m k), respectively, ρ is the density (Kg/m^3) and c_p is specific heat (J/m^3k).

The reduction rates as shown in **Figure 4** were 15.52%, 20.88% and 2.31%, 2.80% for the addition of nano-Cu_2O, nano-Al_2O_3 at concentrations of 1.0 and 3.0 wt.%, respectively, in comparison to pure paraffin. According to the decrease in thermal conductivity and also thermal diffusivity for nano-Cu_2O, nano-Al_2O_3 at 5.0 wt.%, the specific heat increased to 2.427 J/m^3k, 3.081 J/m^3k, respectively which it equals 2.1739 J/m^3k for pure paraffin. As shown in **Figure 4** the peak reduction percentage is 48.43%, compared to pure paraffin in the case of hybrid nanoparticles at 5.0 wt.%. Since the specific heat capacity of nano-Cu_2O is lower than that of nano-Al_2O_3 by about 13.52%, 18.60% and 21.23% at 1.0, 3.0 and 5.0 wt.%, respectively. The same conclusion is revealed by Kok [32]. The author integrated paraffin wax as PCM with alumina (Al_2O_3) and copper oxide (CuO). His results showed that copper oxide has a lower specific heat capacity than alumina. Also, the influence of the occupied volume variation of nanoparticles immersed into the paraffin was studied by Sushobhan and Kar [33]. Their results agree with the present work since the specific heat of the composites decreased by increasing the volume fraction of nanomaterials through the composite.

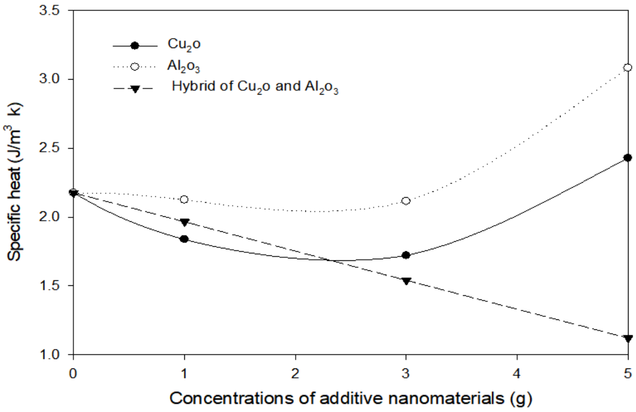

Figure 4. The specific heat of the NEPCMs at different concentrations of 1.0, 3.0 and 5.0 wt.%.

From the above results, the hybrid nano-Cu_2O-Al_2O_3 paraffin wax composite at concentration of 5 wt.% is the best among the studied NEPCMs in this work for thermal energy storage system because it has the highest thermal conductivity and diffusivity. It also has the lowest specific heat, which is considered a good feature in this case as it quickly reaches the melting point.

4. Conclusions

In this study, high thermal conductive nano-additives materials were used to prepare NEPCMs by adding different concentrations of Cu_2O, Al_2O_3 and a mixture of the two oxides in a ratio 1:1 into the paraffin wax. In general, adding high thermal conductive nano-additives increases the thermal conductivity and thermal diffusivity of the paraffin nano-oxide composites while decreasing the specific heat in comparison to pure paraffin. The peak values of thermal conductivity and diffusivity are achieved at a wight concentration of 3.0% when nano-Cu_2O, nano-Al_2O_3 are added to paraffin wax with significant superiority for nano-Cu_2O. While both of those thermal properties are negatively affected by increasing the concentration beyond this value. The results also showed the excellence of the proposed hybrid nanoparticles compared to nano-Cu_2O, and nano-Al_2O_3 as they achieved the highest values of thermal conductivity and diffusivity at a weight concentration of 5.0 wt.%. It also has the lowest specific heat among the studied samples at a weight concentration of 5.0%. Increasing the weight concentration in the case of hybrid nanoparticles may be led to more improvement in the thermal properties. So, it is strongly recommended for future work to study the proposed hybrid nanoparticles paraffin wax at higher weight concentrations.

Author contributions: Conceptualization, SMS, AAES and MRIR; methodology,

SAE; software, ARES; formal analysis, NS; investigation, NS; data curation, NS; writing—original draft preparation, NS; writing—review and editing, SMS; visualization, ARES; supervision, AAES, SAE, MRIR, SMS; project administration, AAES. All authors have read and agreed to the published version of the manuscript.

Acknowledgments: This paper is based upon work supported by Science, Technology and Innovation Funding Authority (STIFA), Capacity building project, Code: 42922.

Conflict of interest: The authors declare no conflict of interest.

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