

CARBON NANOTUBES IN PASSIVE COOLING

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Introduction

Phase change materials (PCMs) have received a great interest nowadays in many practical applications such as energy storage, thermal protection, and active and passive cooling of electronic devices. The phase change of PCMs falls into the category of moving boundary problems. During phase change process, both liquid and solid phases are present and they are separated by a moving interface. For using phase change materials in active and passive cooling applications, the PCMs should possess high thermal conductivity to meet the required heat exchange rates. Moreover, these PCMs should have a small volume change and little degrees of supercooling. However, it is a challenging nowadays to find the ideal PCM that satisfies all the desirable properties [1]. Paraffin waxes are used as PCMs for thermal storage applications because of their desirable characteristics, such as high latent heat of fusion, negligible supercooling, low vapor pressure in the melt, chemically inert and stable, self nucleating, no phase segregation and commercially available at low cost. However, waxes exhibit some inherent limitations, such as low thermal conductivity and large volume change during phase transition. When paraffin waxes are used in energy storing systems, its lower thermal conductivity reduces the heat exchange rate during melting and solidification cycles and therefore the overall power of the phase change regenerator. In the case of space based power systems, this represents a great issue because of a large amount of heat should be stored or dissipated in a short period of time [1]. As reported by Bugaje [3] the phase change (melting or solidification) time is the most important design parameter in latent heat storage systems.

The lower thermal conductivity of paraffin waxes can be enhanced by using metal fillers, additives and/or fins [2, 3]. In the case of aluminum additives [3], it was found that the phase change time reduced significantly in heating and cooling processes. However, the metal fillers add significant weight and cost to the storage systems. Erk and Dudukovic [4] utilized low-density additives such as porous silica

catalyst while Chapotard and Tondeur [5] used activated carbon. They have shown how the mean pore size of the additives could be of critical importance on the system performance. If it is too small, the PCM molecular motion will be hindered thereby it will be very difficult to impregnate the porous media with the PCM which will affect adversely on the latent heat storage capacity. Conversely, if the pores are too large, there will not be sufficient capillary force to retain the liquid wax. For activated carbons, waxes are well retained in micro-pores but they lose their phase change properties. Pay et al. [6] reported that mesoporous activated carbons would perform better but they added that, those two porous supporters (Silica catalyst and activated carbon) do not present real improvement of the effective thermal conductivity.

Carbon fibers have a strong resistance to corrosion and chemical attack. Thus, they are supposed to be compatible with most PCMs. The thermal conductivities of carbon fibers are considerably high. In addition, the densities of carbon fibers are less than 2260 kg/m^3 , which is lower than those of metals that are usually used as additives. Fukai et al. [1] investigated the effect of using carbon fibers as additives. They have used two sets of fiber architectures. First, fibers were randomly distributed in the paraffin waxes. Second, fibers were oriented parallel to the heat flow. The transient temperature response and the effective thermal conductivity of the composites were measured. They concluded that in the case of random fiber distribution, the fiber length has little effect on thermal performance. Using brush type carbon fibers significantly enhanced the effective thermal conductivity of the systems [1]. Mauran et al. [7] used a solid matrix made of graphite as a support for low thermal conductivity reactive salts. This support presented several advantages such as achieving high external heat transfer coefficient, very low bulk density, good mechanical properties, and chemical inertness. Graphite matrix is known to be superior to porous metallic foams made of aluminum, copper or nickel. Pay et al [6] proposed a supported PCM made of paraffin impregnated by capillary forces in a compressed expand natural graphite matrix. They found that the thermal conductivity of the composite equal to that of the sole porous graphite matrix. Moreover, the composite presented the same anisotropy with respect to the compression axis. They concluded that, the compressed expanded natural graphite matrix induced a decrease in the overall solidification time and achieved more stabilization of the thermal storage power.

As another solution for overcoming the undesirable PCMs characteristics, a mixture of different PCMs has been used. Jianfeg et al. [8] studied experimentally the charging process of a cylindrical heat storage capsule filled with stearic acid, sliced paraffin and lauric acid as phase change materials. The geometric structure of the

capsule made up of three coaxial red copper tubes. The surface temperature of the outer surface of the outer tubes is kept at constant temperature by putting it in a constant temperature water bath. The results showed that the using of multiple PCMs enhances the charging rate of the PCM storage.

In this paper a new enhancement technique of the thermal performance of the paraffin wax as a PCM is introduced. Two types of carbon nanotubes with different mass ratios were dispersed into the paraffin wax. The transient temperature response for each mixture was measured during solidification process to predict and calculate the cooling rate and different thermal properties of each mixture. An analytical model was performed to predict the effective thermal conductivity for the new nanocomposites.

Experimental setup

Several samples were prepared by dispersing carbon nanotubes with different mass ratios (1, 2, 3, and 4%) with the paraffin wax using wetting and melting technique. Two types of carbon nanotubes with different surface characteristics were used (surface treated and surface untreated). For the sake of comparison, a pure paraffin wax sample was used also. The total mass of each sample was 20 grams. All Nanocomposite samples were put in cylindrical glass enclosures of 2 cm in diameter and 2.5 cm in length. The enclosures were insulated from their tops and bottoms and the heat is allowed to propagate only in the radial direction. The experimental setup as shown in Fig.(1) consists of a well-controlled furnace equipped of 10-molybdenum disilicide heating elements. These elements are uniformly spaced in a full circle around the interior furnace wall to achieve a uniform temperature distribution. The furnace is a vertical tube/bottom-loading furnace with a working volume of 9" in diameter and 11.5" high modified to accept a tube of 6" O.D. An airlift mechanism, pneumatic system, with top and bottom plugs is supplied to the furnace to attain a variable ambient to the test samples. To attain a variable heat flux, a programmable controller for the input power to the furnace power leads is provided. The transient temperature distribution for each sample under test is measured by means of multiple K-type thermocouples. A Data Acquisition System is used to convert the thermocouples output to a PC. A standard RS-232 asynchronous serial communication is used to connect the Multimeter Data Acquisition System with the PC.

All samples were introduced simultaneously and uniformly distributed into the furnace core, which attained a uniform heat flux. After sufficient duration time for melting, the pneumatic air system was activated to bring down the samples from the furnace core. The transient temperature response for each sample was recorded during

the solidification process simultaneously.

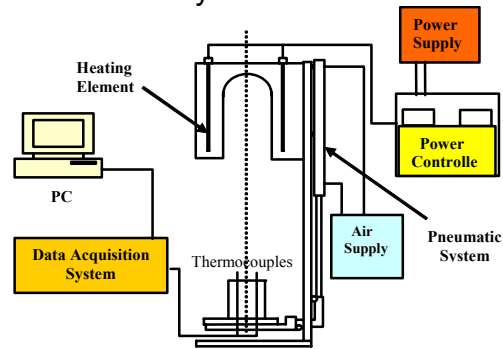


Fig. (1) Experimental Setup

Results and Discussion

To perform a comparative study for the thermal behavior of each sample during solidification process, a solidification characteristic curve for each one of them was established; figures (2-7). Each figure represents the Temperature-Time history for each sample during its solidification process. The melting temperature of the pure paraffin wax is about 67 °C. As shown in the figures, one can observe that the duration time during the phase change process, around the melting temperature, is much greater than sensible heat part. Particularly, the duration time during the phase change process decreases with the increase of the CNT ratio, which means that the cooling rate increases with the increase of the CNT ratio. This in turn will enhance the thermal performance of the paraffin wax system.

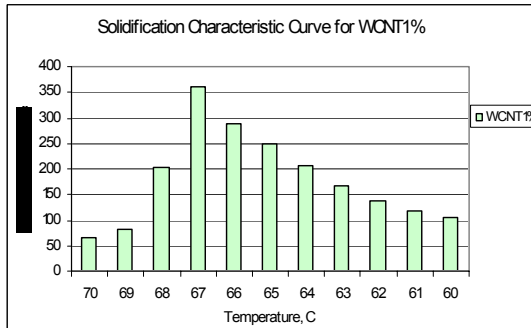


Figure (2)

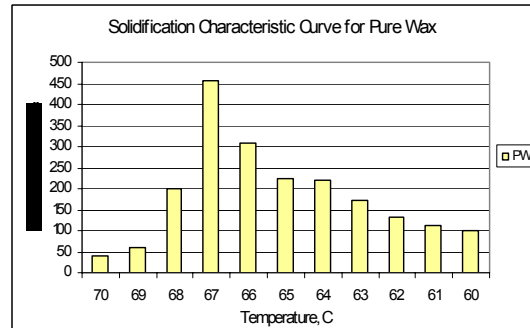


Figure (3)

For all samples, densities and thermal diffusivities were measured (Figures 8 and 9). A Laser Flash Technique was employed to measure thermal diffusivity measurements. In this technique, a very short and uniform laser pulse is directed to a small thickness sample. The pulse usually lasts less than one millisecond. The radiation energy of the pulse is absorbed at the front surface which results in a temperature rise on the back surface of the sample. A thermal detector measures this temperature rise as a function of time. The thermal diffusivity of the sample in the direction perpendicular to the front

and back surfaces can be determined by measuring the thickness of the sample, L , and the half time $t_{1/2}$ which is the time required for the temperature of back surface to reach 50% of its maximum value ($\alpha_{\perp} = 0.139L^2/t_{0.5}$). Figure (9) shows thermal diffusivity as a function of sample with various mass percent of CNT. As shown in the figure, the thermal diffusivity increases with increasing of CNT ratio into the paraffin wax.

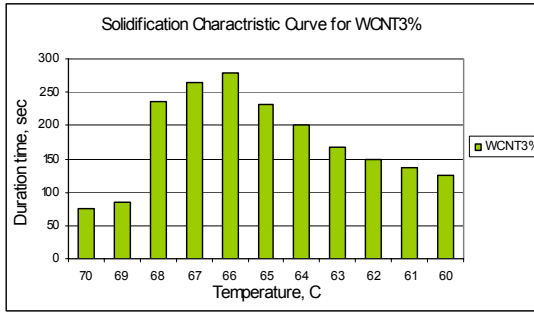


Figure (4)

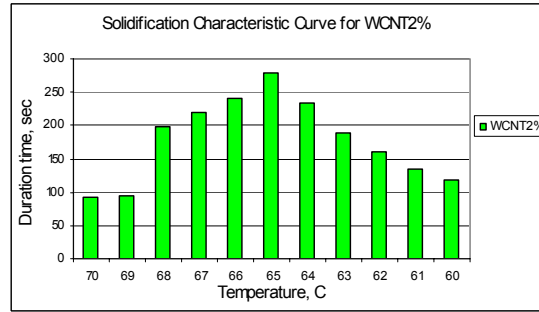


Figure (5)

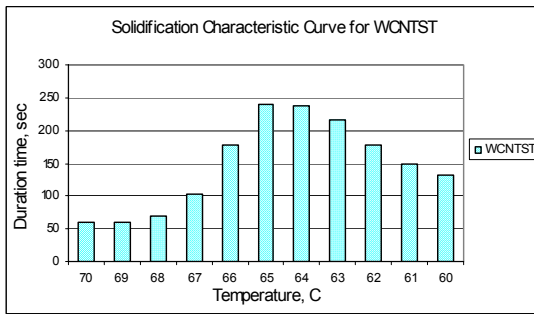


Figure (6)

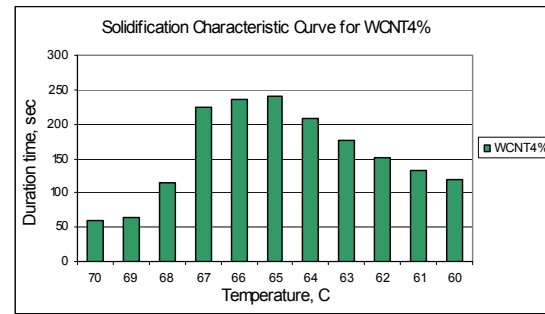


Figure (7)

The thermal diffusivity of pure paraffin wax is about $0.00151 \text{ cm}^2/\text{sec}$ while the thermal diffusivity of the paraffin wax filled with 4% CNT is about $0.00268 \text{ cm}^2/\text{sec}$. This corresponds to an increase in performance of about 44%. However, the thermal diffusivity of Wax-surface treated carbon nanotubes based nanocomposites is $0.0028 \text{ cm}^2/\text{sec}$ (about 47% performance increase).

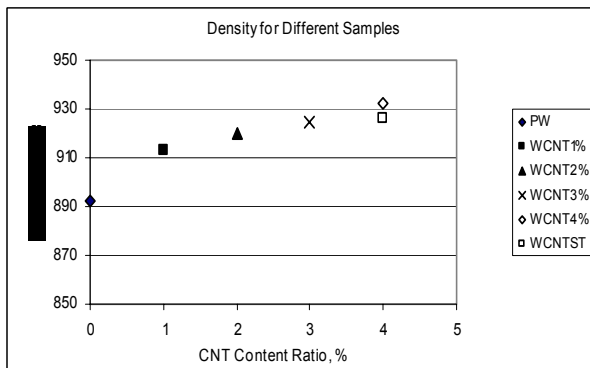


Figure (8)

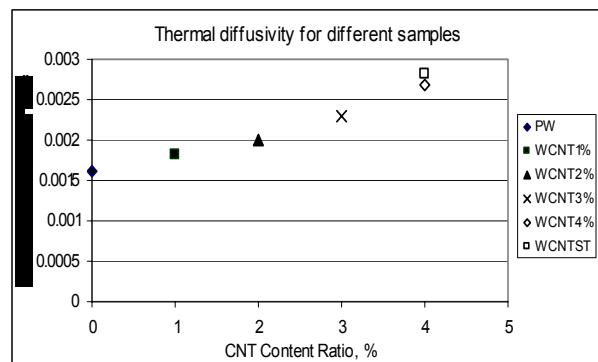


Figure (9)

The heat capacities of pure wax and nanocomposites were measured by Differential Scanning Calorimetry technique and thermal conductivities were calculated. As shown in Figure 10, specific heat decreases by increasing CNT ratio, which means that wax-CNT nanocomposites displayed less ability to store heat but high aptitude to conduct heat. However, the thermal conductivity increases by increasing the CNT mass percent (Figure 11). As a result of that, the cooling rate increases by increasing the CNT content as shown in Figure (12). The enhancement of the cooling rate was about 40% in the case of 4 % mass ratio.

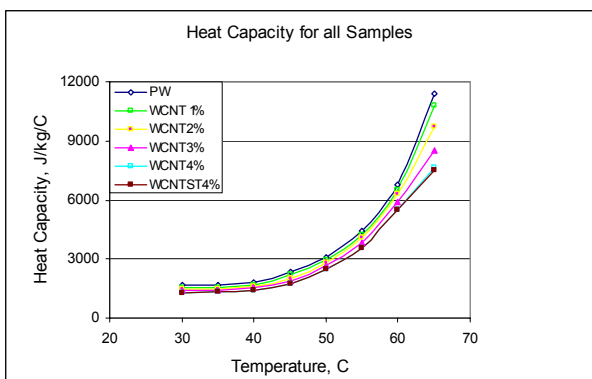


Figure (10)

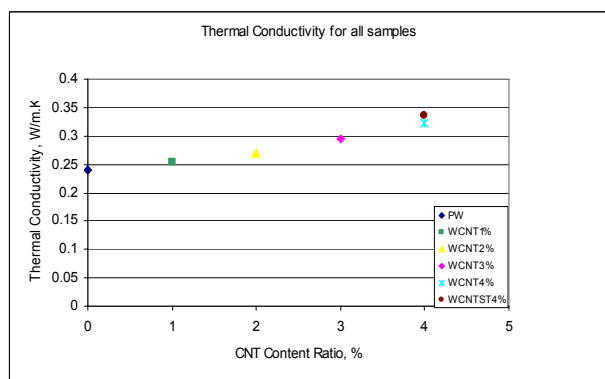


Figure (11)

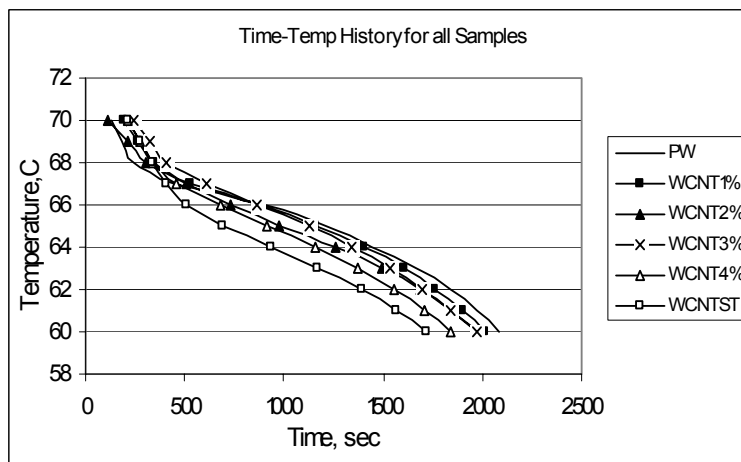


Figure (12)

Analytical model to predict the effective nanocomposites thermal conductivity

An analytical model was performed to predict the effective thermal conductivity of CNT based wax nanocomposites. The analytical solution was based on one dimensional heat conduction approach for a composite matrix cell consists of a low thermal

conductivity PCM in which carbon nanotubes were dispersed. It is assumed that, the carbon nanotubes were distributed into the PCM in a regular scattered distribution under the following assumptions:

- 1- The CNTs have the same outer diameter, D_o , inner diameter, D_i , and length L . Also we assume that $D_i = D_o/2$.
- 2- The CNTs are homogeneously distributed into paraffin wax as indicated into figure (13), where the CNTs are perpendicular to each other and separated by a distance $D_o/2$ from each side and the bulk paraffin wax has a pore diameter of ($d_p = L + 2 D_o$).
- 3- For a symmetry consideration we will take the half of the indicated cell.
- 4- The unit cell will be divided into three layers, I, II, and III. The first layer, (I) has a length of $D_o/2$, and consists of paraffin wax and CNTs; the second layer, (II) has a length of $D_o/2$ and consists of pure paraffin wax; and the third layer, (III) has a length of $(d_p - 2 D_o)/2$, figure (14).
- 5- The thermal conductivity in all directions should be the same due to the symmetry of the geometry in all directions.

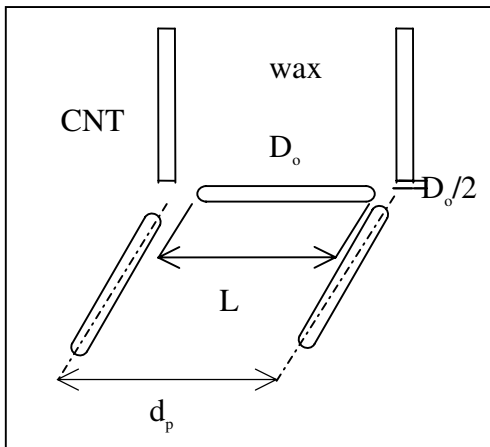


Figure (13) CNTs distribution

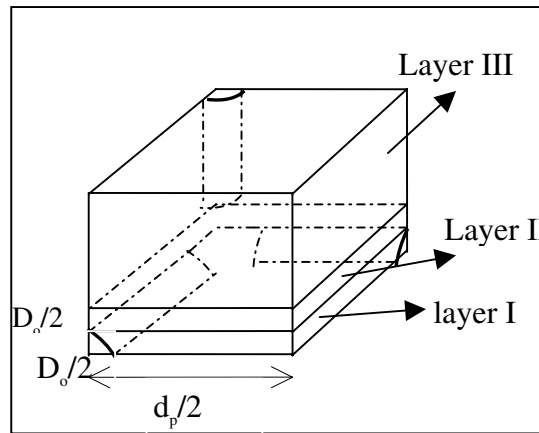


Figure (14) Physical Model Unit Cell

Layer (I):

The total volume of layer (I), $V(I) = \frac{d_p^2}{8} \cdot D_o$

Estimation of the content ratio of each component at layer (I)

The solid volume ratio:
$$\frac{V_s}{V(I)} = \frac{3\pi}{8} \left(\frac{D_o}{d_p} \right) \left(1 - 2 \left(\frac{D_o}{d_p} \right) \right)$$

The Polymer volume ratio:
$$\frac{V_p}{V(I)} = \left(1 - \frac{\pi}{2} \left(\frac{D_o}{d_p} \right) \right) \left(1 - 2 \frac{D_o}{d_p} \right)$$

The air volume ratio:
$$\frac{V_a}{V(I)} = \frac{\pi}{8} \left(\frac{D_o}{d_p} \right) \left(1 - 2 \frac{D_o}{d_p} \right)$$

Layer (II): As mentioned above layer (II) consists of pure paraffin wax and its total volume is: $V(II) = \frac{d_p^2}{8} \cdot D_o$ and $\frac{V_p}{V(II)} = 1$

Layer (III): The total volume of layer (III), $V(III) = \frac{d_p^2}{8} \cdot (d_p - 2D_o)$

Estimation of the content ratio of each component at layer (III):

The solid volume ratio:
$$\frac{V_s}{V(III)} = \frac{3\pi}{16} \left(\frac{D_o}{d_p} \right)^2$$

The polymer volume ratio:
$$\frac{V_p}{V(III)} = \left(1 - \frac{\pi}{4} \left(\frac{D_o}{d} \right)^2 \right)$$

The air volume ratio:
$$\frac{V_a}{V(III)} = \frac{\pi}{16} \left(\frac{D_o}{d_p} \right)^2$$

The Effective Thermal Conductivity for each layer:

By considering K as the effective thermal conductivity for each individual layer; this property depends on the geometry and volume ratios of the contents.

Effective Thermal Conductivity for Layer (I):

By introducing $\left(\frac{D_o}{d_p} \right) = X$, we can find that:

$$K(I) = \frac{V_s}{V(I)} \cdot K_s + \frac{V_p}{V(I)} \cdot K_p + \frac{V_a}{V(I)} \cdot K_a$$

$$K(I) = \left(\frac{3\pi}{8} \cdot X \cdot (1 - 2X) \right) \cdot K_s + \left(1 - \frac{\pi}{2} \cdot X \cdot (1 - 2X) \right) \cdot K_p + \left(\frac{\pi}{8} \cdot X \cdot (1 - 2X) \right) \cdot K_a \dots\dots(1)$$

Effective Thermal Conductivity for Layer (II):

$$K(II) = 1 \cdot K_p = K_p \dots\dots\dots(2)$$

Effective Thermal Conductivity for Layer (III):

$$K(III) = \left(\frac{3\pi}{16} \cdot X^2\right) K_s + \left(1 - \frac{\pi}{4} \cdot X^2\right) K_p + \left(\frac{\pi}{16} \cdot X^2\right) K_a \dots\dots\dots(3)$$

The Effective Thermal Conductivity of present nanocomposites, K_e :

$$\left(\frac{L_I + L_{II} + L_{III}}{K_e}\right) = \left(\frac{L_I}{K_I}\right) + \left(\frac{L_{II}}{K_{II}}\right) + \left(\frac{L_{III}}{K_{III}}\right)$$

$$\left(\frac{d_p/2}{K_e}\right) = \left(\frac{D_o/2}{K_I}\right) + \left(\frac{D_o/2}{K_{II}}\right) + \left(\frac{(d_p - 2D_o)/2}{K_{III}}\right) \text{ , which leads to}$$

$$\frac{1}{K_e} = \frac{X}{K_I} + \frac{X}{K_{II}} + \frac{(1-2X)}{K_{III}}$$

$$\frac{1}{K_e} = \frac{X \cdot (K_{II}) \cdot (K_{III}) + X \cdot (K_I) \cdot (K_{III}) + (1-2X) \cdot (K_I) \cdot (K_{II})}{(K_I \cdot K_{II} \cdot K_{III})} \text{ or, in other words}$$

$$K_e = \frac{(K_I \cdot K_{II} \cdot K_{III})}{X \cdot (K_{II}) \cdot (K_{III}) + X \cdot (K_I) \cdot (K_{III}) + (1-2X) \cdot (K_I) \cdot (K_{II})} \dots\dots\dots(4)$$

Where; K_I , K_{II} , and K_{III} , are calculated from equations (1), (2), and (3) respectively.

Equation (4) predicts the effective thermal conductivity of the present nanocomposites as a function of the CNT outer diameter and the paraffin wax pore diameter, which can be introduced in terms of volume fraction of the Carbon nanotubes.

Model Validation

The effective thermal conductivities for the present nanocomposites were predicted by utilizing equation (4). Figure (15) shows a good agreement between the analytical and experimental findings which reflects the validity of the present analytical model.

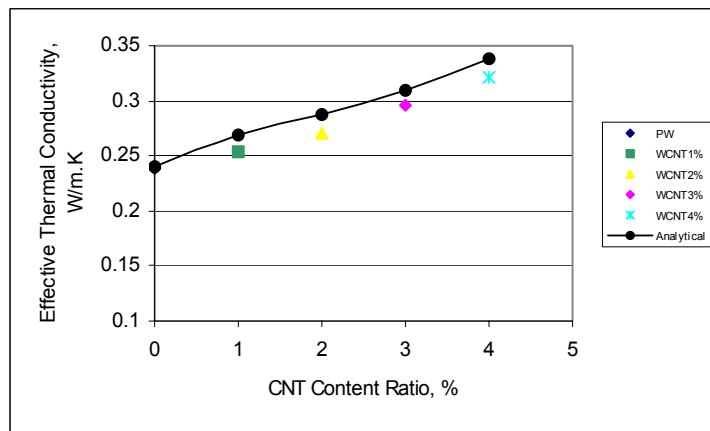


Figure (15) Analytical Model Validation

Conclusions

Carbon nanotubes were employed to enhance the thermal properties of paraffin wax. Two types of carbon nanotubes with different surface characteristics and different mass ratios were dispersed into the paraffin wax. The nanocomposite thermal performance was enhanced significantly by increasing the mass ratio of the CNT into the paraffin wax. As a result of this enhancement, the cooling rate during the solidification process increases significantly by increasing the mass ratio of the CNT. An analytical model was performed to predict the effective thermal conductivity of nanocomposites. The analytical and experimental findings show a good agreement, which reflects the validity of the predicted analytical model.

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