

Research Paper

Tuning Thermal Properties of Beeswax Incorporating ZnO, CuO and Soot Particles

Dinsefa Mensur Andoshe*, Brook Essay, Osman Ahemd, Anduaem Merga, Demeke Tesefaye, Nahom Gezahegn, Tariku Miressa and Tamirat Merid

Department of Materials Science and Engineering, School of School of Mechanical, Chemical and Material Engineering, Adama Science and Technology University, P. O. Box 1888, Adama, Ethiopia

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Abstract

Phase change materials that have low thermal conductivity take a longer time to store and to discharge the stored thermal energy. The thermal conductivity of phase change materials can be enhanced using different additives. Beeswax is a phase change material with a high thermal capacity and low thermal conductivity. Therefore, the thermal conductivity, melting temperature, latent heat, and heat capacity of beeswax incorporating ZnO, CuO and soot particles with the 0.0, 0.064, 0.326, 1.304, and 3.260 g in the 20 g beeswax have been studied. The thermal conductivity measurement shows that the incorporation of ZnO, CuO and Soot particles improved the thermal conductivities of the beeswax, i.e., from 0.2502W/mK to 2.89W/mK. Moreover, the differential scanning calorimetry measurements revealed that the melting temperature, latent heat, and heat capacity of the beeswax are decreased because of the incorporation of ZnO, CuO and Soot particles. Therefore, it is possible to tune the thermal properties of the beeswax incorporating metal oxide, soot particles to use beeswax-metal oxide/soot composite as a phase change materials.

1. Introduction

Phase Change Materials (PCMs), as thermal energy storage systems, is receiving greater attention in recent decades for applications in heating and cooling. The PCMs substances undergo a change in phase; solid to liquid or liquid to gas or vice-versa, they absorb large amounts of heat while maintaining constant temperature and discharge the absorbed heat when it needed. Thermal energy can be stored either as a sensible heat (SH) or latent heat (LH). Thermal energy is stored as SH while heating/cooling a liquid or solid storage medium. The amount of SH stored depends on the specific heat of the medium, the temperature change, and the amount of storage material (IoanSarbu et al., 2018; Ioan et al.,

2019). A medium that stores SH in which the materials have a large temperature rise/drop when storing/releasing thermal energy. Materials that store thermal energy as LH are PCMs and they release or absorb the thermal energy while their physical state is changed. LH is mainly stored during the phase-change process in the isothermal condition and then it is related with the latent heat of the substance. LH storage system using PCMs is an effective way to store thermal energy and has the advantages of high energy storage density and isothermal nature of the storage process. The stored LH can then be discharged at the desired rate which coincides with the melting temperature of the PCMs.

* Corresponding author, e-mail: dinsefadear@gmail.com

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However, the main disadvantage of LH system is its low thermal conductivity, which mostly falls in the range of 0.2 W/(m·K) to 0.7 W/(m·K); therefore, effective heat transfer enhancement technologies must be adopted (IoanSarbu et al., 2018). Among the PCMs materials beeswax is paraffin obtained from bees with the molecular formula of $C_{15}H_{31}COOC_{30}H_{61}$ (Sara et al., 2015; Lakshmi et al., 2016). The melting temperature of beeswax is in the range of 61-67°C (Muhammad et al., 2017). The variation of the melting temperature of beeswax may be attributed to the impurities.

Beeswax has been studied for PCMs; for example, it is used as a PCM material to maintain at an ambient temperature of the operating photovoltaic panels at high solar radiation and buildings (Muhammad et al., 2017; Thaib et al., 2018). The photovoltaic panel surface operating at high solar radiation has a surface temperature close to 49°C. However, when PCM material is applied on the surface of the solar panel, the surface temperature reduces to 34°C and the efficiency of the solar panels improves. The drawbacks of PCMs materials are their low thermal conductivities that affect the rate of thermal energy storage, and the PCM materials exhibited a large melting temperature. Usually, to improve the thermal conductivities of PCM materials, a metal oxide or metal particles have been incorporated/added in the PCMs. The thermal properties of beeswax with addition of grapheme and CuO have been studied in previous work and the result showed that the thermal conductivity of the beeswax is enhanced (Nandy et al., 2016; Muhammad et al., 2017; Chengjun et al., 2019).

Therefore, this article focus on tuning the thermal properties of beeswax by incorporating different metal oxide particles and comparing the obtained thermal properties of beeswax/metal oxide composites. The thermal properties of beeswax, such as thermal conductivities, heat capacities, and melting temperature, are studied incorporating ZnO, CuO, and soot particles. Furthermore, the metal oxide and soot particles also analyzed using XRD to study the crystal structure and phase.

2. Materials and Methods

2.1. Materials used

The study used beeswax as its PCM material, along with $Zn(NO_3)_2$, $CuSO_4$, NaOH, Soot, deionized water

and ethanol to synthesize metal oxide particles. The precursor used in this study were $Zn(NO_3)_2$ with purity of 96%, $CuSO_4$ and NaOH with purity of 99.8 % which were purchased from central drug house (p) Ltd India and Ranchem industry, respectively. Moreover, the thermal properties of beeswax/metal oxide composite were characterized by DSC-60 plus and QTM-500. Furthermore, the crystal and phase of metal oxide particle were investigated using XRD-7000.

2.2. Experimental procedure

2.2.1. Preparation of ZnO, CuO and Soot Particles

ZnO particles were synthesized by precipitation method using NaOH and $Zn(NO_3)_2$ precursors (Romadhan et al., 2016). The NaOH and $Zn(NO_3)_2$ solution was prepared by adding 2.0 g of NaOH in 50 ml and separately 14.875 g of $Zn(NO_3)_2$ precursor in 100 ml of deionizing water. Both beakers containing the solution were heated up to 80°C with continuous stirring kept at 1000 rpm in temperature controlled magnetic stirrer for an hour. After the complete dissolution of $Zn(NO_3)_2$ and NaOH in de-ionized water, the mixture was transferred to a single beaker and kept at constant stirring rate of 1000 rpm for two hours keeping the temperature at 80°C. The precipitated solution formed after two hours reaction was separated using a centrifuge to obtain ZnO and the obtained residue was washed several times with deionized water until all the traces of NaOH were removed. Then, the ZnO gel were dried in an oven at 250°C for about an hour.

The CuO particles were prepared using the same methods with a bit modification from (Nandy et al., 2016) with a precursor of $Cu(SO_4)$ and Na(OH). The 16.0 g of $Cu(SO_4)$ was dissolved in 100 ml of distilled water and 4 g of NaOH was dissolved in 150 ml of distilled water putting the beakers on the magnetic stirrer adjusting the string rate to 1000 rpm. After complete dissolution of $Cu(SO_4)$ and Na(OH) in the DI water the solutions was transferred to a single beaker drop by drop under constant stirring condition. Final solution reaction was allowed to proceed for 2 hr, then after the solution was allowed to settle for an overnight and the supernatant solution was then discarded carefully. The precipitate was washed several times using distilled water to remove residual from the obtained CuO. Finally, the obtained CuO gel was dried in oven for overnight at 80°C. The Soot consisting of

agglomerated particles was collected from regular car exhaust.

The brown color beeswax was purchased from Adama city, Ethiopia, and repeatedly washed to remove the dusts and impurities that may encounter during the collection and/or transportation. It was known that beeswax is insoluble in water and washing does not affect it except removing the impurities.

2.2.2. Preparation of Beeswax-Metal oxide Particle Composite

Beeswax-metal oxide composite was synthesized by mixing a 20 g of beeswax with 0.0, 0.064, 0.326, 1.304, and 3.260 g of metal oxide particle using the ultrasonic device. The detail compositions of beeswax and metal oxide are presented in Table 1. The Beeswax-metal oxide composite was synthesized with the following procedure. The beeswax sample was melted at 66°C, which is a bit higher than the melting temperature of the beeswax, for 60 min. Then, the synthesized metal particle using the precipitation method was added to the melted beeswax sample. The mixing of beeswax and metal oxide particle performed using an ultrasonic vibrator with a frequency of 40 kHz at a temperature of 66°C to keep the sample in a liquid state. The vibration of the beeswax-metal oxide composite using ultrasonic sonicator continued for 2 hr., until the particles and beeswax mixed evenly. Then, the melted beeswax-metal oxide composites were cast and let to dry in ambient condition; the obtained beeswax-metal oxide composites are shown in Figure 2. The synthesized beeswax-metal oxide composite show a slight color changes comparing with bare beeswax. The observed color change is from brown to light brown for ZnO incorporation, from Brown to light green for addition of CuO and from brown to black as shown in Figure 2(a)-(n).

Table 1: Amount of particles incorporated in the beeswax

S. No	Beeswax(g)	Particles(g)
1	20	0.064
2	20	0.326
3	20	1.304
4	20	3.260

3. Results and Discussion

The synthesized metal oxide using precipitation methods and the collected soot powder from regular

automobile were characterized by XRD. Moreover, their crystallite size also calculated using Scherrer’s formula as shown in equation 1. The obtained XRD data were matched with JCPDS48, 1548 for CuO, and JCPDS 36, 1451 for ZnO sample and they are polycrystalline as observed that the incident x-ray have diffracted from different planes as shown in Figure 1. The synthesized CuO and ZnO also found to be monoclinic and wurtzite phases, respectively. The (-111) and (111) plane peak intensity of CuO is higher than the intensity other planes, which suggested that the dominant growth direction could be (-111) and (111). The XRD spectra suggesting that the incident x-ray diffracted more on the (101), (100) and (002) planes from the wurtzite ZnO crystal, and the growth direction also dominated to (101) direction. Moreover, the average crystallite size for the synthesized CuO and ZnO found to be about 30 nm and 35 nm, respectively.

$$d= K\lambda(\beta \cos \theta)^{-1}..... (1)$$

Where “d” is the mean size of the crystallite/grain size, K is constant with the value about 0.9, λ is X-ray wave length, β is the line broadening at half the maximum intensity (FWHM) and θ is Bragg angle.

The XRD spectra of soot black powder, which is collected from car exhaust, is presented in Figure 1(c). Soot powder collected from different source is composed of a crystalline graphitic carbons (Manoj et al., 2012; Pradip et al., 2012). The peak position exactly resembles with the work of Manoj et al. (2012). The crystallite size estimated using Scherrer’s equation is about 40 nm.

3.1. Thermal Conductivity and Melting Temperature

The measured thermal conductivities for the prepared beeswax-metal oxide composites are shown in Figure 2. Measurement of thermal conductivity of beeswax-metal oxide composites were conducted using quick thermal conductivity measuring (QTM). The thermal conductivities of the Beeswax enhanced after CuO and ZnO particles were incorporated, however, the Soot particles have slightly decreased the thermal conductivities of the beeswax. Moreover, the amount of thermal conductivities enhancement of the beeswax also depend on the amount of metal oxide particle added in the beeswax matrix.

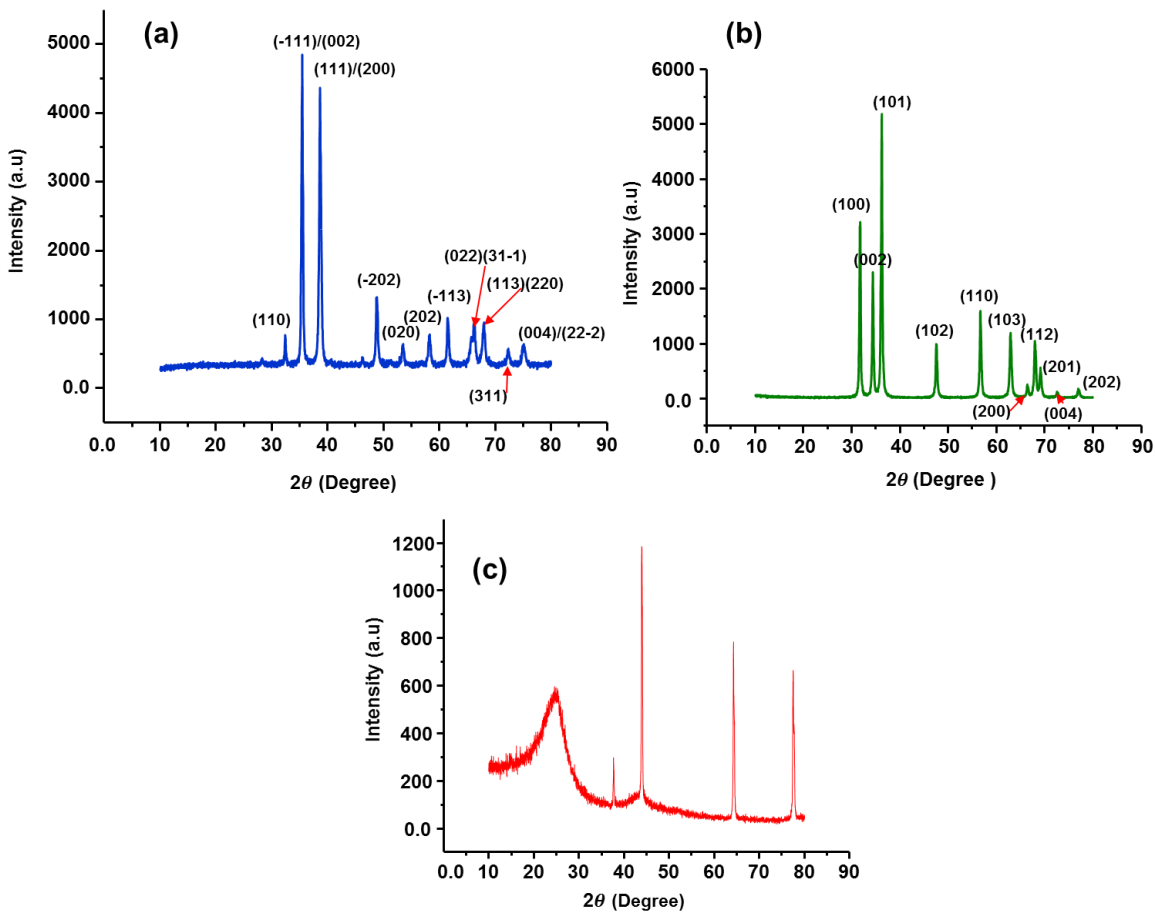


Figure 1: XRD spectra of (a) CuO, (b) ZnO, and (c) Soot powder collected from regular car exhausts

More specifically, the thermal conductivities of beeswax after incorporation of 0.0, 0.064, 0.326, 1.304 and 3.260 g of ZnO particles became 0.2502, 0.45, 1.90, 2.5 and 2.89 W/mK, respectively. Furthermore, after 0.0, 0.064, 0.326, 1.304 and 3.260 g of CuO particles are added to the 20 g of beeswax the thermal conductivities were measured as 0.2502, 0.4903, 2.221, 3.1004, and 3.9552 W/mK, respectively. Moreover, after incorporation of 0.064, 0.326, 1.304 and 3.260 g of soot particle to the 20 g beeswax matrix, the thermal conductivities were measured as 0.2502, 0.3525, 0.7520, 1.3132 and 1.749 W/mK, respectively as shown in Figure 3. Therefore, it is possible to deduce that the thermal conductivity of the beeswax can enhance by the addition of ZnO, CuO and Soot particles.

3.2. Melting Temperature, Latent Heat and Heat Capacity

The latent heat, heat capacity and melting temperature of beeswax-ZnO, beeswax-CuO and

beeswax-Soot composite with different mass metal oxide particle were measured using DSC. The DSC measurement reveals that addition of ZnO and CuO particles has decreased the melting temperature, latent heat and heat capacity of beeswax, however, the soot particles is enhanced the melting temperature, heat capacity and latent heat of the beeswax as shown on Figure 4 and Figure 5. More specifically, the measured melting temperatures of the beeswax-metal oxide composite were 65°C, 63.1°C, 62.2°C, 62.1°C and 61.8°C, when the mass of CuO particle were 0.0, 0.064, 0.326, 1.304 and 3.260 g, respectively. The ZnO particle also decreased the melting temperature of beeswax from 65°C to 63.2°C, 62.6°C, 62.5°C and 62.4°C when 0.0, 0.064, 0.326, 1.304 and 3.260 g is incorporated in the 20 g beeswax matrix, respectively.

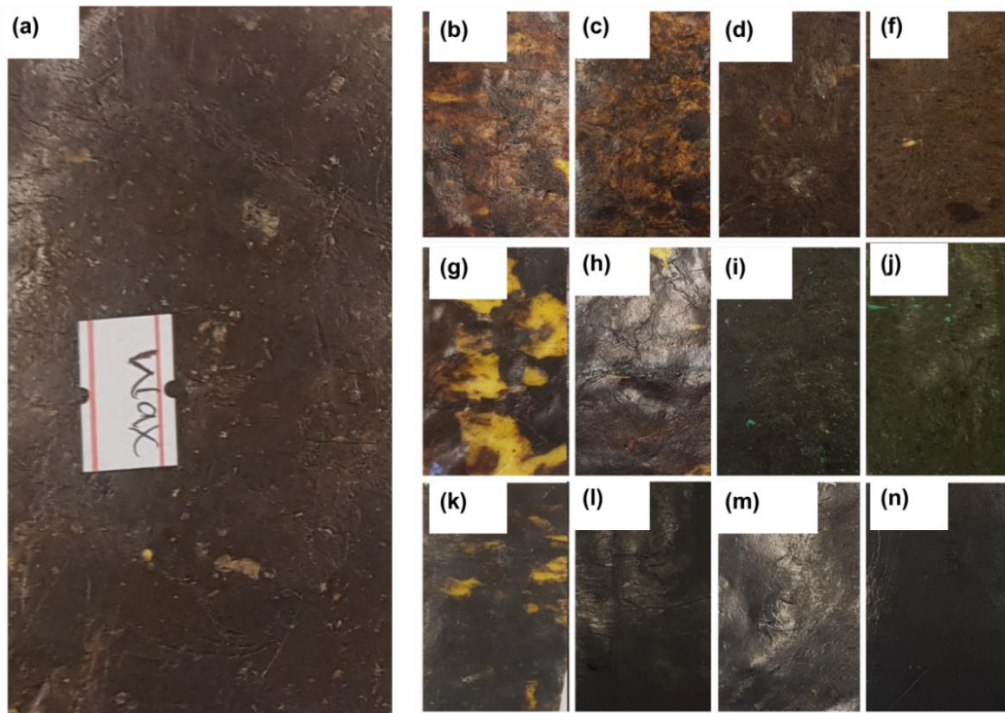


Figure 2: The beeswax-metal oxide composite with their thermal conductivity, (a) bare beeswax, (b)-(f) 20 g beeswax with 0.064, 0.326, 1.304 and 3.260 g ZnO composite respectively, (g)-(j) 20 g beeswax with 0.064, 0.326, 1.304 and 3.260 g of CuO, respectively and (k)-(n) 20 g beeswax with 0.064, 0.326, 1.304 and 3.260 g of Soot, respectively.

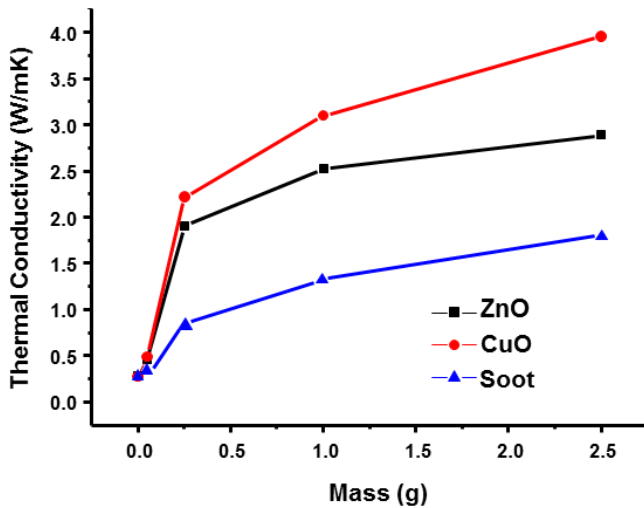


Figure 3. Measured thermal conductivities of beeswax-metal oxide composites vs. mass.

When soot particles with 0.0, 0.064, 0.326, 1.304 and 3.260 g are incorporated in the 20 g beeswax matrix, the melting temperatures were measured as 65°C, 64.8°C, 64.6°C, 64.2°C and 63.8°C, respectively. After the incorporation of metal oxide with the mass of 0.0, 0.064, 0.326, 1.304 and 3.260 g the latent heat of beeswax-metal

oxide composite also recorded as 400, 350, 195, 125 and 105 KJ/Kg for CuO, respectively; 400, 360, 340, 270 and 187.5KJ/Kg for ZnO, respectively and 400, 395, 360, 350 and 300 KJ/Kg for soot, respectively as shown in Figure 4(b).

Figure 5 shows that when the heat capacity of beeswax-metal oxide composite with different mass of metal oxide (ZnO and CuO) increased, the heat capacity of the beeswax decreased. On the other hand, for soot particle, the heat capacity is enhanced when the amount of added mass in the beeswax matrix is increased. The heat capacity for beeswax-CuO composite is recorded as reduced to 2.301, 1.75, 1.23, 0.63 and 0.25 KJ/Kg°C for incorporation of 0.0, 0.064, 0.326, 1.304 and 3.260 g of CuO particles, respectively. Furthermore, the heat capacity of beeswax-ZnO composite was decreased (from 2.301) to 2.119, 2.035, 1.242 and 0.994 kJ/kg°C for 0.064, 0.326, 1.304 and 3.260 g of ZnO particles, respectively. Moreover, the heat capacity of beeswax-Soot composite was also decreased (from 2.301) to 2.281, 2.175, 2.533, 2.0 and 1.75 kJ/kg°C for 0.064, 0.326, 1.304 and 3.260 g of soot particles, respectively.

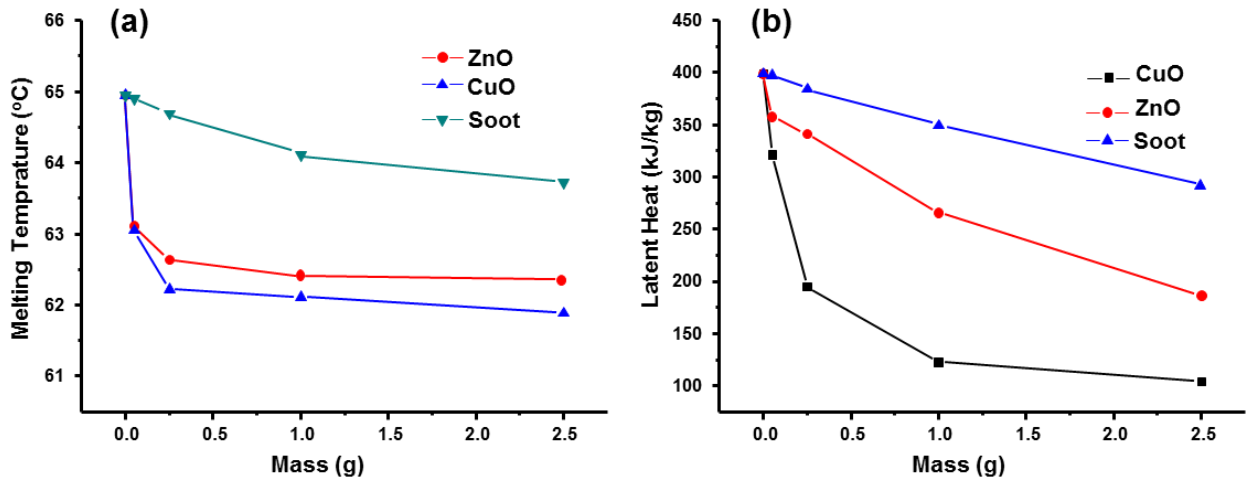


Figure 4: Melting temperature and latent heat of beeswax-metal oxide composite. (a) Melting temperature of beeswax-metal oxide composite vs. different mass of metal oxide particles (b) latent heat of beeswax-metal oxide composite vs. different mass of metal oxide particles.

of CuO particles, respectively. Furthermore, the heat capacity of beeswax-ZnO composite was decreased (from 2.301) to 2.119, 2.035, 1.242 and 0.994 kJ/kg°C for 0.064, 0.326, 1.304 and 3.260 g of ZnO particles, respectively. Moreover, the heat capacity of beeswax-Soot composite was also decreased (from 2.301) to 2.281, 2.175, 2.533, 2.0 and 1.75 kJ/kg°C for 0.064, 0.326, 1.304 and 3.260 g of soot particles, respectively.

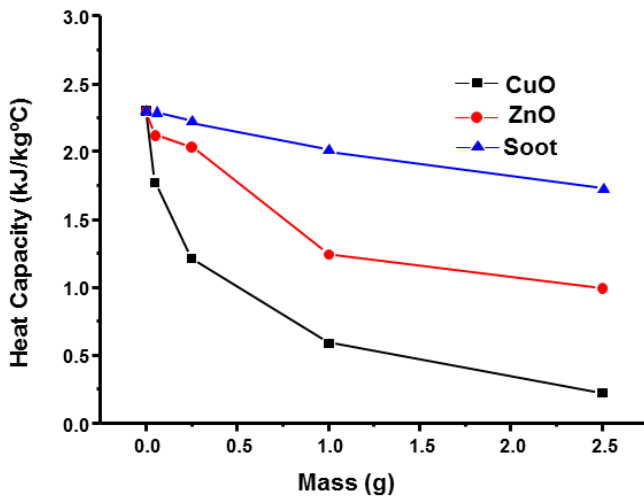


Figure 5: Heat Capacity of beeswax-metal oxide composite.

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