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High performance Thermal Coating Comprising (CuO:NiO) Nanocomposite/C Spectrally Selective to Absorb Solar Energy

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ABSTRACT

A novel nanocomposite consisted of nanomaterials as (CuO:NiO) and carbon (fuel ash) were designed to absorb solar energy. Thin films were made via casting and spin coating of the dopants nanocomposite thin films, containing different concentration ratios of CuO:NiO. These thin films are precipitated on a glass and copper substrates. The optical properties of the doped fuel ash films with nanoparticles were measured in the range of 250-1300 nm. The intensity of solar radiation was measured too. The data were analyzed and interpreted in terms of the theory of phonon-assisted direct electronic transitions. The E_g of the doped C was measured with different concentration ratios of (CuO:NiO) (A=0.5:2.5, B=1:2, C=1.5:1.5, D=2:1, E=2.5:0.5) wt. %, with a fixed concentration of C of (7) wt.%. The results of the doped samples revealed an energy gap of (2.5-3.9 eV) and the absorptivity ranged from (85-99 %) for all nanocomposites. The energy gap of this nanocomposite system is very similar to those of semiconductor and has high efficiency to absorb the solar energy radiation. In addition, the results showed that the heat absorbed by the samples subjected to solar energy on the surface would be a selective surface. Thus, the synthesized coating will be utilized on a flat plate collector as a trap to absorb solar energy. Prog. Color Colorants Coat. 13 (2020), 275-284© Institute for Color Science and Technology.

1. Introduction

Most regions in Iraq have around 250 days of strong sunlight in a year and the energy per square meter in a day may be around 5 kWh [1]. In order to harvest solar energy, nanocomposites were coated on a flat plate collector. This approach allows the coated collector to be a spectrally selective surface to absorb solar energy and hence, reduce the emission of the solar energy to efficiently convert the sunlight to heat [1]. Optical properties are most important for many applications in protective coatings on flat plate collectors, microelectronic, and optoelectronic devices depending on the properties of reflectance and absorbance for the thin films during their preparation. Doping the materials, such as carbon, with nanoparticles can cause a remarkable change in dopants properties [2, 3].

Copper oxide (CuO) is an important material because of its many potential applications and the availability with abundance in nature. Thereby, it has main phases of semiconductor and structure of the monoclinic crystal. CuO has high ionization energy and low formation energy, which accelerate the incorporation of Cu into the lattice and can create localized states in the band gap with NiO to dope

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carbon. It has prominent activator in nanocomposite thin films [4, 5]. Additionally, nickel oxide (NiO) extensively used in photo-catalyst and semiconductors applications, due to its anti-ferromagnetic properties, wide band gap and highly ordered crystal structure make it possible to form a thin films [6, 7]. Emissivity is an important benefit in technological applications and scientific studies. It is an important thermo-physical factor describing the capability of the surface to emit thermal radiation. Spectral emissivity measurement at low temperatures was inspected with an experimental apparatus for the samples. The spectral emissivity of alumina sample with high-purity (99 wt. %) was measured by an advanced algorithm to remove disturbances through background radiation and was calculated by the least-square method [8, 9]. The spectral emissivity calculated for the sample is estimated to be less than 5.1% for the spectral range considered [10-12]. Spray coating technique was used to deposit nickel oxide on cleaned glass substrates, the energy band gap was found to be in the range of 3.64 and 3.86 eV [13-15].

Herein, we present an investigation on the effect of doping C (fuel ash) with (CuO:NiO) nanoparticles as a mixture with different concentration ratios to improve the physical properties of carbon. To the best of our knowledge, there is no study on the optical properties of (CuO:NiO/C) was published until now. The accurate determination of the optical constants for such materials is important, not only to know the basic behavior but also to develop and exploit their features in technological applications.

2. Experimental

Nanocomposite thin film materials were prepared via spin coating and casting methods, in which the nanocomposite was precipitated on the copper (Cu) and glass substrates. Carbon ash was obtained from Doura Refinery as an output of the combustion process. It was used after several processes carried out on it such as sieving to eliminate all Lingering things. The SEM test was utilized to find out the particle size which was found in the range of (29.62-93.22 µm). The chemical composition of the carbon ash is shown in Figure 1 by EDS inspection.

The particles of nanomaterials CuO and NiO were obtained from the company Changsha Easchem /Hunan-China, the particle size for them was 50 nm and purity 99.9 %. AFM technique was used to check the particle size of the nano-materials as shown in Figure 2. The AFM results showed that the particle size was 58.43 and 30.69 nm for CuO and NiO, respectively. Figure 2 shows the AFM images for CuO and NiO nanoparticles.

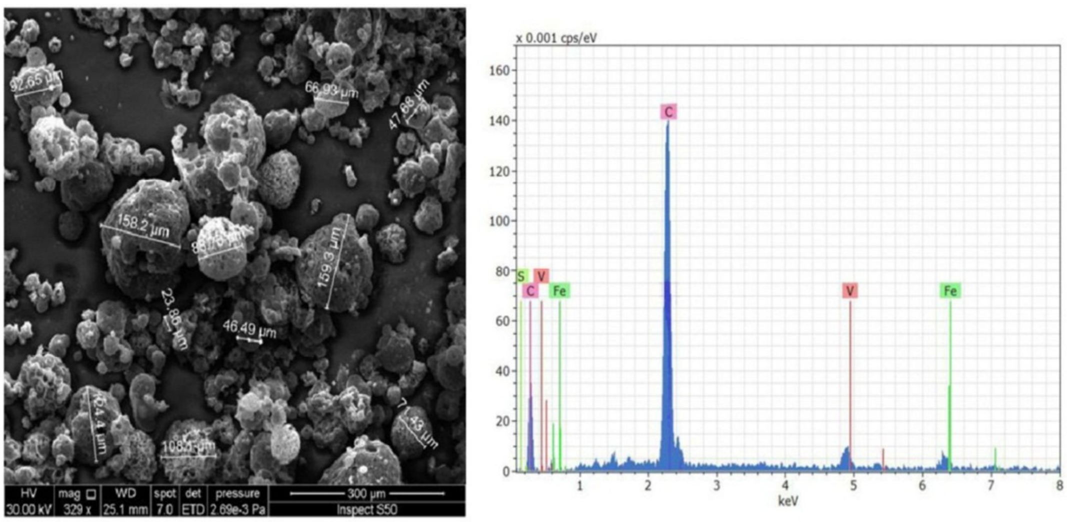

Figure 1: SEM and EDS images of carbon (fuel ash).

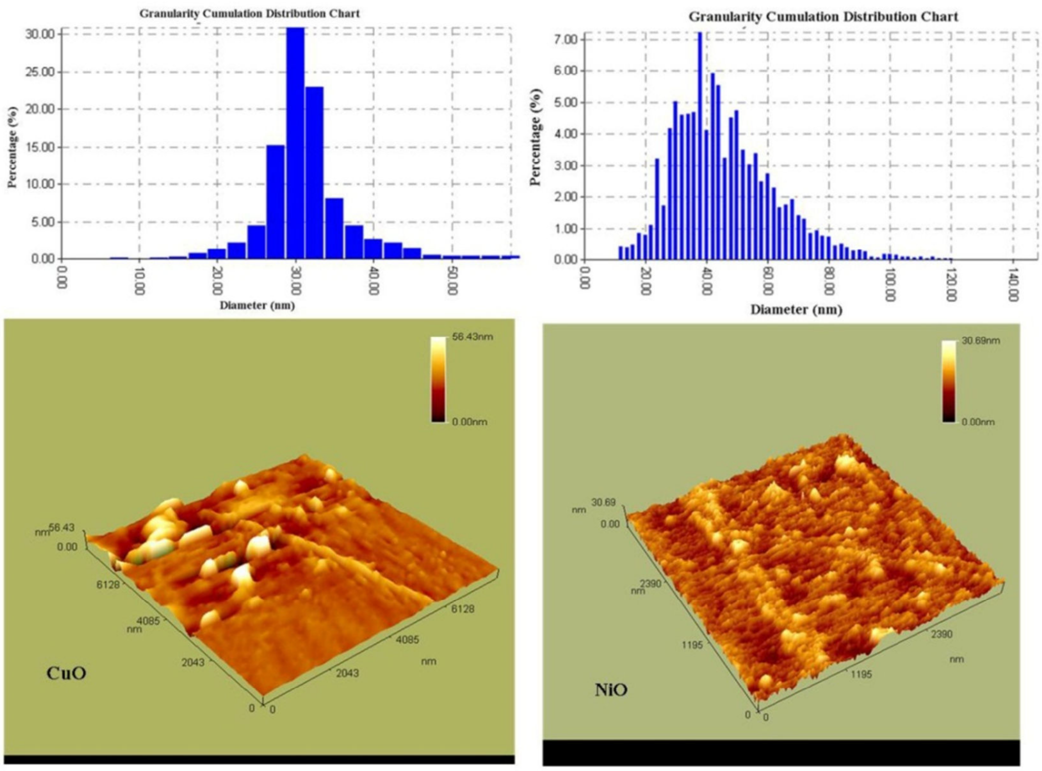

Figure 2: AFM images for CuO and NiO nanoparticles.

2.1. Synthesis of nanocomposite

The composite was prepared via mixing stoichiometric amounts of the CuO and NiO nanoparticles with the carbon (fuel ash). An electronic balance (5 digits) was used to weigh the desired amount of the compositions. The composite was prepared in different concentration ratios in which the amount of carbon (fuel ash) was fixed at 7 wt %. The CuO:NiO were added in different weight ratios (A=0.5:2.5/7, B=1:2/7, C=1.5:1.5/7, D=2:1/7, and E=2.5:0.5/7) wt. % of the colloidal. The mixture was blended in the deionized water and molasses followed by stirring for 3 h using ultrasonic to give a homogeneous mixture.

The colloid was transferred to a glass tube and followed by vigorous shaking before film deposition. The films were deposited on pre-cleaned copper and glass substrates with dimensions of (2.5 × 5 cm^2). Prior to the deposition process, the substrates were completely rinsed several times using acetone and ethanol to clean the surface and to remove any grease. Finally, the substrates were rinsed in deionized water and dried in the oven at (50 ^oC) for 1 h. The nanocomposite materials were deposited on substrates via spin coating and casting methods and dried in the oven at (100 ^oC) to remove any residual solution in samples. The thickness of the films was measured using the SEM device and it was found to be around 300 nm.

2.2. Characterization

The prepared samples were analyzed using computerized diffused reflectance, Avantes DH-S-BAL-2048. UV-Vis Spectro-2048 was used to record the wavelength in the range of (250-1300 nm). The absorbance spectra were recorded using computerized SHIMADZU, UV-VISIBLE SPECTROMETER UV-1650 PC in the wavelength range from (200-800 nm). All optical investigations were performed at room temperature (300 K).

2.3. Measurement of solar energy and heat absorbed

The intensity radiation of solar energy was measured via data logger (daystar type DS-05) in W/m^2. The data collected through direct exposure of samples to sunlight. The temperatures values were measured using digital reader type HT-9815-XINTEST with thermocouple type K (-200~1372 ^oC). The thermocouples were fixed on the backsides of the samples using sticky tape and by a thermal silicon bar. The coated samples fixed on a small table with a tilt angle of 45^{o}, these samples tilted to the sun radiation toward the south latitude 33o and longitude 44o representing the experiment location in Iraq. The changes in temperature were recorded using the digital data logger to calculate the heat absorbed by the coating over the Cu substrate. Figure 3 shows the coated samples on the stage provided with a data logger during the exposure to sunlight.

3. Results and Discussion

3.1. Optical properties studies

Optical properties study of the nanocomposite thin films (CuO:NiO/C) plays a significant role in understanding the nature of optoelectronic. These properties can be interpreted in the viewpoint of the interference between the incident solar radiation and the semiconducting properties of the films, and through the study of the band gap energy obtained from optical absorption by explaining the band structure of semiconductors [16, 17]. The investigation and discussions of optical properties of the nanocomposite thin films (CuO:NiO/C) depend on measurements of reflectance, R(λ), in the spectral range (250-1300 nm). These optical studies utilized to determine the optical energy gap (E_g) for the nanocomposite thin films.

3.1.1. Reflectance spectra

The reflection, R(λ), spectra of the investigated samples (A, B, C, D, and E) of the nanocomposite thin films (CuO:NiO/C) are shown in Figure 4. It is clear that, these spectra are divided into two special regions: (i) the high absorption region represented in the region of wavelength ≤ 420 nm and (ii) the less absorption region, which lies between (420-1000 nm). Furthermore, as the concentration of (CuO:NiO) increases, the reflectance becomes relatively higher. This is maybe attributed to increasing of CuO concentration and decreasing NiO concentration, as appeared in the reflectance curves in the Figure 4, which has a high absorption nature than NiO. All samples exhibited a good absorbance for all constitutes of the nanocomposite concentrations. Sample E gave the best absorption value of about (98 %) Also, the absorption edge is shifted toward the lower energies as (CuO) concentration increases [18, 19]. This result confirms that the incident light on the nanocomposite films was thoroughly absorbed, which provides good evidence that the surfaces of the deposited thin films have roughness nature to harness the incident light in visible to near IR region. The absorptivity (α) can be calculated using the relation between the reflectivity (R) and the absorptivity (α) for all nanocomposite samples. the results showed that the absorptivity (α) varies in the range of (88-98 %) due to change in nanoparticle concentrations.

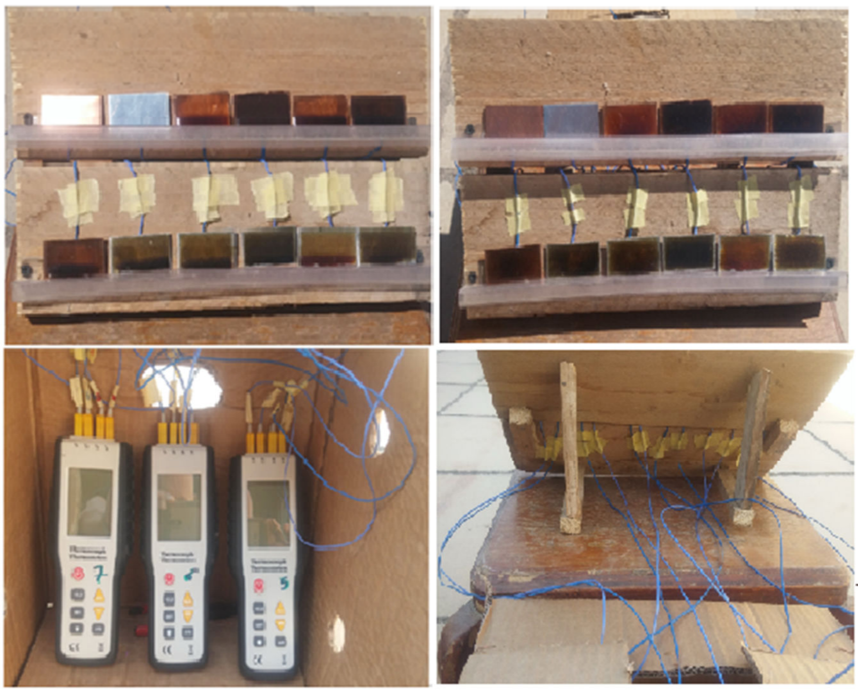

Figure 3: samples with thermocouples and data loggers to measure the temperatures.

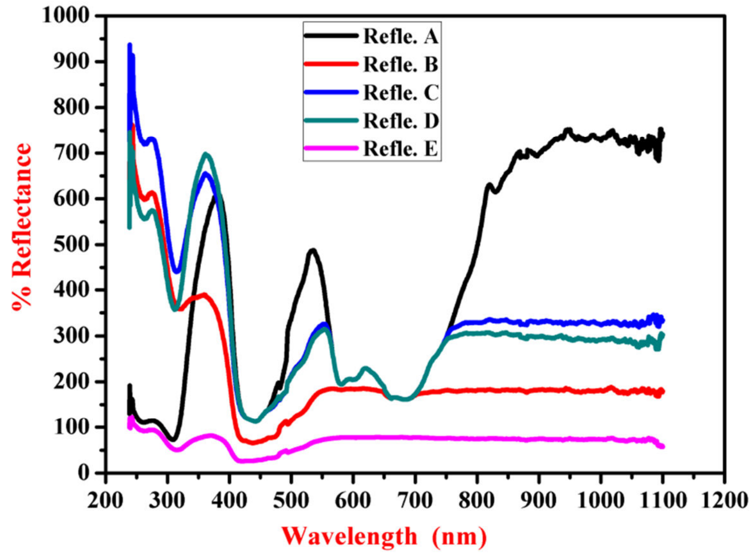

Figure 4: reflectance test for all concentration of nanocomposite samples.

3.1.2. Optical band gap

Based on the results obtained by the reflectivity measurements, the Kobelka Munk equation was used to calculate the energy gap for the nanocomposite thin films for samples, which is almost similar to Tauc Relation. The standard equations (Eq. 1 and Eq.2) are:

$$(F(r)*h\nu)^{1/n}=A(Eg-h\nu) \ldots \quad (1)$$

Where F(r) is:

$$F(r)=\frac{k}{s} \ldots\ldots \quad (2)$$

Where, F(r) Kobelka Munk function, ($h\nu$) is the photon energy, (E_g) is the energy gap for the material, (A) is a constant of band tailing parameter, (k) is the molar absorption coefficient, (s) is the scattering factor and (n) is the power factor for the transition mode. It is dependent upon the nature of the material, whether it is crystalline or amorphous and the photon transition. n=2 for direct transition and n=1/2 for the indirect transition.

Figure 5 reveals two values of energy gap for concentrations (A, C, D, and E) and three values of the concentration (B) for the doped carbon by (CuO:NiO). The energy gap values varied between (3.1-3.7 eV) for the concentrations (A and C) which may be attributed to higher content of NiO. On the other hand, concentrations (D and E) may be attributed to CuO content. The energy gap values of (B) between (2.5-3.9 eV) are attributed to NiO concentration in the nanocomposite thin films. The results of energy gap are highly consistent with previous research [20, 21]. The values of energy gap were calculated according to the above equations. The energy gap values for all concentrations of the samples (A, B, C, D, and E) are listed in the Table 1 as follow:

Table 1: Energy gaps values for all samples.

E_g A (eV)	E_g B (eV)	E_g C (eV)	E_g D (eV)	E_g E (eV)
3.1	2.5, 3.0	3.1	3.1	3.1
3.7	3.9	3.7	3.7	3.7

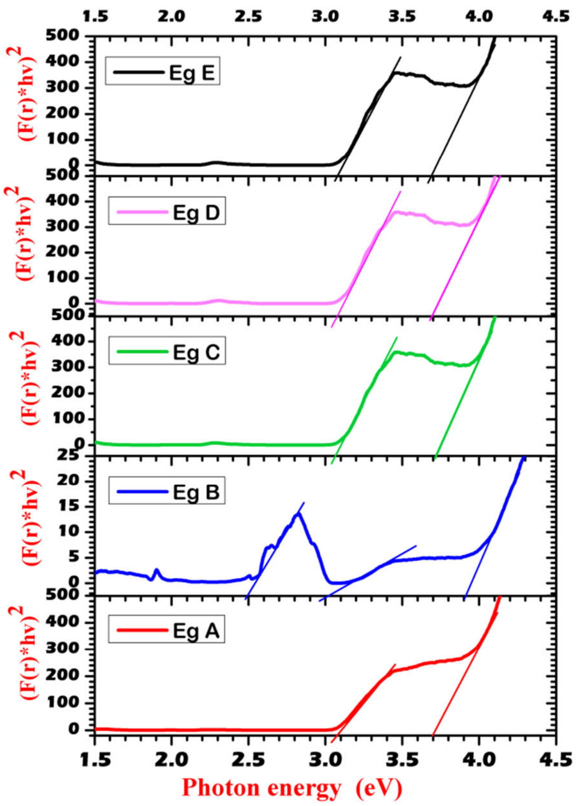

Figure 5: energy gap for all nanocomposite thin films (CuO:NiO/C) of the samples.

The absorption edge for the particles was shifted with a variation of the concentrations towards higher or lower energy, these values prove that the nanocomposite thin films acted as semiconductor, and revealed high absorption for the incident light of the solar energy. Sample E (2.5:0.5/7 wt. %) showed the best coating nature for the surface of the thin film, this attributed to the high content of CuO in the thin film composition. Thus, solar radiation absorption is significantly affected by the roughness of the coated surface. The surface roughness improves the capture of solar energy because it doesn't permit the solar energy to reflect through it and be absorbed thoroughly on the surface coating. The energy gap (E_g) values exhibited good values after doping compared with other values in the researches [20-24].

3.2. Heat absorb from solar energy

Heat absorbed calculation was done by subjecting the coated samples to solar energy directly. After measuring the temperature, the absorbed heat can be calculated using the following equation (Eq. 3) [17, 18]:

$$Qabs. = mc\frac{\Delta T}{\Delta t} \quad (3)$$

Where, Q is the heat absorbed (w/m^2), m is the mass of the sample (kg), c is the specific heat for the copper (J/kg k), T is the temperature (k) and t is the time (s). Figure 6 shows that the heat absorbed varied with the time of samples exposure through the day; the absorption peak was in the noon time. The absorptivity and emissivity data were recorded for the coated plate of all samples, and compared with data obtained from another plate without coating used as an internal standard.

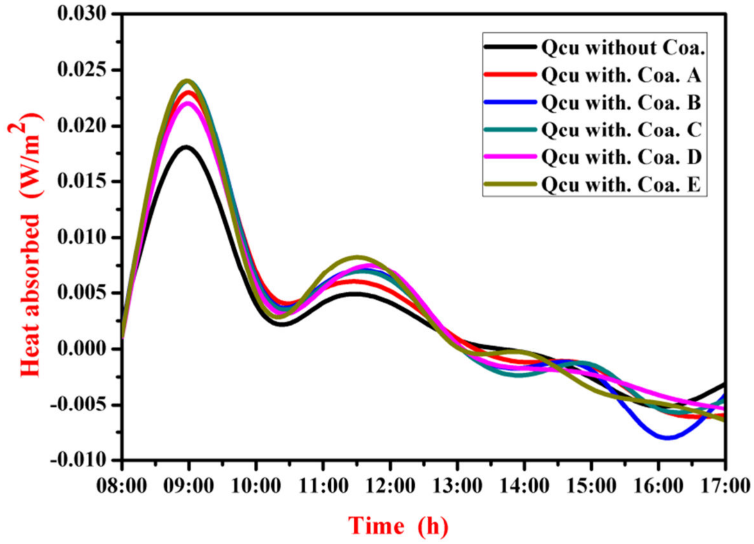

Figure 6: heat absorbed through samples with one sample without coating.

The heat radiation calculated via the following equation (Eq. 4) [17, 18];

$$Qrad = \sigma\left({T_1}^4 - {T_2}^4\right) \quad (4)$$

Where, Q is the heat radiation (w/m^2), σ is the Stefan-Boltzmann constant (w/m^2 k^4), and T is the temperature in (K) between the plates from the first plate to the second plate. These values of Cu features are used in calculations shown in Table 2 such as [23-28].

Figure 6 shows the best concentration was (E=2.5:0.5/7 wt. %) which has high absorptivity, that obtained from the absorption of the solar radiation and has the best surface coating of the thin films over the substrate. Figure 7 shows the variation of temperature and the incidence radiation of solar energy with the time along the day. The peak value is appeared from figure at the noon time [21-27].

Figure 7 shows the higher temperature absorbed of the coated samples over the surface of sample E that was revealed a good agreement with the previous results, which has the highest absorption rate and lower value of emissivity.

The emissivity was calculated from the relation between the heat absorbed and the heat radiated for the coated samples in comparison with the non-coated one. The variation of emissivity with the time is shown in Figure 8 for all coated and non-coated samples. the emissivity for all samples decreases with the rise of temperature; this depends on the morphology of the coated surface and its ability to absorb the solar energy. The roughness of the coated surface prevents the reflection of solar energy and increase the absorptivity of the thin films. Figure 8 proved that the best cluster for the thin film coating was over the sample E (2.5:0.5/7 wt. %), which has the best combination of CuO and NiO to dope carbon (fuel ash). the highest absorbance ranges of the solar energy radiation exhibit the values around (85-99 %). These values of the absorptivity coincided with the theoretical values that are obtained from the reflectivity test [9, 24-32].

Table 2: values of Cu features.

Specific Heat (C) KJ/Kg. K	Density (ρ) kg/m^3	Stefan Boltizman constant (σ) w/m^2 k^4	Conductivity (k) w/m k
0.385	8945	5.67 E-8	386

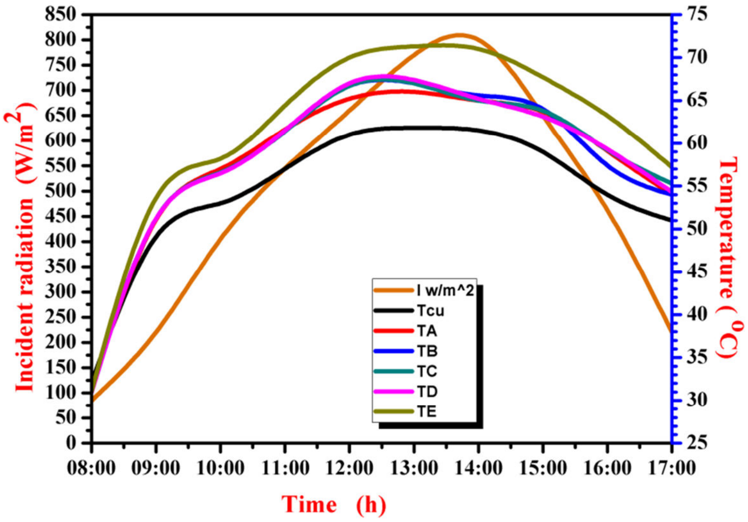

Figure 7: Incidence radiation and temperatures with time.

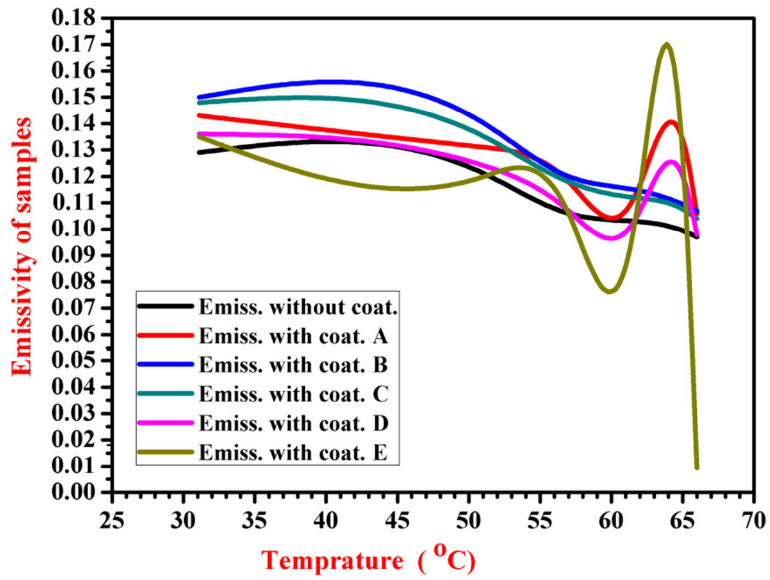

Figure 8: variation of emissivity with temperatures without and with coating.

4. Conclusion

The nanocomposite material of thin films was deposited on the copper and glass substrates with different concentrations. The ratios of (CuO:NiO) varied with a constant concentration of carbon (fuel ash). The deposition process was done via spin and casting methods, which give a high quality coating. The optical energy gap of the coating layer in the UV-Visible and NIR region of the nanocomposite films were calculated. The energy gap was influenced by the doping type of metal complexes, which utilized in the coating layer. The values of energy gaps depended on the concentration of the CuO and NiO in the mixture and their ability to dope carbon. Therefore, the values of the energy gaps were in the range of (2.5-3.9 eV). The reflectivity results supported the high absorptivity (α) measurements for the incident light, which appeared in the values range (88-98 %). A heat calculation depends on the temperatures recorded by the thermocouples. The obtained results showed that the absorbed heat reaches the peak at noon. The absorbance was distributed through the entire coated surface. The temperature distribution throughout the day depends on the incident radiation for solar energy. The emissivity also depends on the temperature and the roughness of the coating layer, when the temperature is high the absorptivity will be high and the emissivity will be lowest, this behavior confirms that these samples are selective surfaces to solar light. Therefore, the high absorptivity and their values are ranged from (85-99 %) which is coinciding between the theoretical and experimental values for the absorptivity.

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