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CARBON-CUPROUS OXIDE COMPOSITE NANOPARTICLES ON GLASS TUBES FOR SOLAR HEAT COLLECTION

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Abstract- Carbon-cuprous oxide composite nanoparticles were chemically deposited on surface of thin glass tubes of spent energy saving lamps for solar heat collection. Carbon was obtained from fly ash of heavy oil incomplete combustion in electric power stations. Impurities in the carbon were removed by leaching with mineral acids. The mineral free-carbon was then wet ground to have a submicron size. After filtration, it was reacted with concentrated sulfuric/fuming nitric acid mixture on cold for 3-4 days. Potassium chlorate was then added drop wise on hot conditions to a carbon slurry followed by filtration. Nanocarbon sample was mixed with 5% by weight PVA to help adhesion to the glass surface. Carbon so deposited was doped with copper nitrate solution. After dryness, the carbon/copper nitrate film was dipped in hydrazine hydrate to form cuprous oxide - carbon composite, It was then roasted at 380-400 °C A heat collector testing assembly was constructed of 5 glass coils connected in series with a total surface area of 1250 cm^2. Heat collection was estimated by water flowing in the glass coils that are coated with the carbon/copper film,. Parameters affecting the solar collection efficiency such as time of exposure and mass flow rate of the water were studied. Results revealed that the prepared glass coil has proven successful energy collector for solar heat.

Key words- Solar collector, nanocarbon, copper, nanoparticles, energy science,

I. INTRODUCTION

Solar heat is one of the best sources of renewable energy with minimal environmental impact. Solar water heating systems can provide nearly all hot water during the summer months and about 50% all year round. The average domestic system can reduce you carbon emissions by about 400kg per year. Michael Arnold [1] reported that an approach that could challenge silicon as the predominant photovoltaic cell material, University of Wisconsin-Madison materials engineers have developed an inexpensive solar cell that exploits carbon nanotubes to absorb and convert energy from the sun. Recent advances have afforded researchers a greater level of control over the chemical makeup of carbon nanotubes, which in turn has opened the door to myriad applications. The thin spaghetti-like tubes are easy and inexpensive to manufacture, stable and durable, and are both good light absorbers and electrical conductors.

Direct absorption solar collectors have been proposed for a variety of applications such as water heating [2] However the efficiency of these collectors is limited by the absorption properties of the working fluid, which is very poor for typical fcuprous oxide used in solar collectors. It has been shown that mixing nanoparticles in a liquid (nanofluid) has a dramatic effect on the liquid thermophysical properties such as thermal conductivity. Nanoparticles also offer the potential of improving the radiative properties of liquids, leading to an increase in the efficiency of direct absorption solar collectors. Here we report on the experimental results on solar collectors based on nanocuprous oxide made from a variety of nanoparticles (carbon nanotubes, graphite, and silver). We demonstrate efficiency improvements of up to 5% in solar thermal collectors by utilizing nanocuprous oxide as the absorption mechanism. In addition the experimental data were compared with a numerical model of a solar collector with direct absorption nanocuprous oxide. The experimental and numerical results demonstrate an initial rapid increase in efficiency with volume fraction, followed by a leveling off in efficiency as volume fraction continues to increase.

Kongkanand et al [3] showed that single wall carbon nanotube (SWCNT) architecture when employed as conducting scaffolds in a TiO_2 semiconductor based photo electrochemical cell can boost the photo conversion efficiency by a factor of 2.

Titanium dioxide nanoparticles were dispersed on SWCNT films to improve photo induced charge separation and transport of carriers to the collecting electrode surface. The shift of ∼100 mV in apparent Fermi level of the SWCNT−TiO_2 system as compared to the unsupported TiO_2 system indicates the Fermi level equilibration between the two systems. The interplay between the TiO_2 and SWCNT of attaining charge equilibration is an important factor for improving photo electrochemical performance of nanostructured semiconductor based solar cells. The feasibility of employing a SWCNT−TiO_2 composite to drive the water photoelectrolysis reaction has also been explored.

Schariff [4] reported that carbon is the most versatile element in the periodic table. Due to its ability to form both sp^3,p^2, and sp hybrids and stable multiple p_π & z.s bnd; p_π bonds, carbon can build up 3-, 2-, 1-, and 0-dimensionally structured substances with a broad variety of physical and chemical properties. In the last decade diamond films, active carbons, carbon fibres, and carbon-carbon composites were extensively studied. The discovery of C_{60} opened up the world of spherical molecular carbon allotropes and gave rise to the development of diverse new materials comprising ultra-hard carbons as well as superconductors. In parallel, the discovery of the carbon nanotubes enabled the synthesis of new absorbents, catalysts and electron emitters. Several new carbon phases, like rectangular diamond or amorphous tetrahedral carbon, are discussed for special applications.

Dillon published in his paper [5] that carbon Nanotubes are fairly suitable for Photo conversion and Electrical Energy Storage. Journet, P. Bernier [6] reported that carbon nanostructures such as single-walled and multi-walled nanotubes (SWNTs and MWNTs) or graphitic polyhedral nanoparticles could be produced using various methods. Most of them are based on the sublimation of carbon under an inert atmosphere, such as the electric arc discharge process, the laser ablation method, or the solar technique. Chemical methods can also be used to synthesize these kinds of carbon materials: the catalytic decomposition of hydrocarbons, the production by electrolysis, and the heat treatment of a polymer, the low temperature solid pyrolysis or the in situ catalysis.

Copper nanoparticles Copper nanoparticles are synthesized from copper sulphate pentahydrate using a novel method. To control the nuclei process, a two-step reduction process is applied. To prevent nanoparticles from oxidization and agglomeration, oleic acid is employed as an extracting and a surfactant. The influence of process parameters (ratio of Cu^{2+} to NaH_2PO_2, pH values, and temperature) on the morphology and dispersion is investigated. All products are cubic phase copper, as determined from X-ray diffraction measurements. Scanning electron microscopy and transmission electron microscopy reveal that the copper nanoparticles are spherical and have sizes of approximately 30 nm. The FT-IR spectrum shows that the copper nanoparticles were coated with oleic acid. Copper metal is used in different applications such as capacitor materials, conductive coatings; Conductive inks; conductive pastes; high thermal conductivity materials; lubricant additives, sintering additives, the superficial conductive coating processing of metal and non-ferrous metal, medicine append material, raw material for bulk nanomaterial. Copper nanoparticles are widely used as catalysts. Efficacious catalyst: copper and copper alloy nanometre featuring high efficacy and selectivity can be used as catalyst in some reactions, e.g. carbon dioxide compound hydrogen to produce methanol. Conductive coatings composite films consisting of metallic Cu nanoparticles dispersed in poly(acrylic acid) (PAA) have been prepared by reduction of Cu^{2+} from the copper salt of PAA above 220 °C under a H_2 atmosphere. Optical absorption properties and structures have also been investigated by UV/VIS, WAXD, TEM and IR. The composite films exhibited an optical absorption peak centred at ***ca.*** 570 nm. The composite film made by heat treatment at 220 °C was less stable because Cu particles in the film were oxidized to Cu^{2+} ions within several weeks, while the composite films prepared by heating above 230 °C were stable and the Cu particles in their films were not oxidized. Shankar et al. [7] suggested that reduction of aqueous $AuCl_4^-$ and Ag^+ ions using extracts from geranium and lemongrass plants occurred due to the ketones / aldehydes groups present in the extract. This plays an important role in directing the shape evolution in these nanoparticles. Printed circuit board (PCB) consists of various metals including precious metals such as copper, gold, silver and palladium [8]. Goosy and Kellner [9] reported the existence of 2% wt of silver in the e-waste. There have been several studies on the recycling of materials by mechanical [10], thermal [11] and chemical [12]-[13] processes, but most of them dealt with recovery of one or two specified materials of all PCB compositions.

Habisreutinger studied the use of carbon/polymer composite in solar heating water systems. [14]. Cheap Tubes Inc [15] reported that carbon nanotubes have different types such as:

- Single Walled Nanotubes-SWNTs
- Double Walled Nanotubes-DWNTs
- Multi Walled Nanotubes-MWNTs
- OH Functionalized Nanotubes-OH-CNTs
- COOH Functionalized Nanotubes-COOH-CNTs
- Short Nanotubes 0.5-2.0um long-Short CNTs
- Short OH Functionalized Nanotubes 0.5-2.0um long-Short OH CNTs
- Short COOH Functionalized Nanotubes 0.5-2.0um long -Short COOH CNTs
- Industrial Grade Nanotubes-IGCNTs
- OH Functionalized Industrial Grade Nanotubes-OH-IGCNTs
- COOH Functionalized Industrial Grade Nanotubes-COOH-IGMWNTs
- Graphitized Nanotubes-GMWNTs
- Carbon Nanotubes Arrays-CNT Arrays
- C60 and other Fullerenes

Fig. 1 Carbon single walled nano tubes (after ref.15)

Nanocarbon tubes are produced by several methods such as arc method, laser method, chemical vapour deposition, and ball milling and diffusion flame method.

The aim of this study is to examine the efficiency of nanocarbon particles doped with cuprous oxide to collect sensible solar heat during rising and lowering the temperature of the day.

II. EXPERIMENTAL

Materials and methods

1. Carbon particles measuring microns was obtained from fly ash of electrical power stations in Cairo Egypt. The mineral was freed from its impurities by leaching with sulphuric acid on hot conditions. It was then filtered, washed with distilled water till free of the acid and dried at 100°C over night.
2. Copper nitrate was of pure chemical supplied by Adwic, Egypt. Potassium chlorate was chemically pure reagent of Adwic, Egypt.
3. Mineral acids of sulfuric and nitric were chemically pure reagents. Water glass was of commercial grade of Awic, Egypt.
4. Bi-distilled water was used for chemical analysis or reactions involved and for measuring the energy collection experiments. Tap water was used for other purposes
5. The glass tube coil was prepared from spent energy saving lamps. It was disassembled manually. The inner coating of the tube was removed by HCl acid followed by blasting of water jet stream. The coil was then coated with carbon nanoparticles slurry in water containing 5% by weight of

PVA as adhesive. The process was repeated to complete coverage. After drying, the carbon-coated glass coil was doped with copper nitrate using super saturated solution of the salt containing 5 % PVA adhesive. The process was also repeated to complete coverage with copper nitrate. The coated coil was then immersed in hydrazine hydrate to reduce the nitrate salt to cuprous oxide. For solar heat collection measurements, 5 glass coils were connected in series and contained in a container of clear cellophane to avoid interference of the surrounding ambient conditions.

6. Size of carbon and cuprous oxide nanoparticles was measured by XRD measurements.

Measurement of solar heat collection efficiency

Efficiency solar heat collection was determined by measuring the temperature of the inlet and outlet of distilled water following through 5 glass coils mounted vertically and connected in series. Figure 1 shows a schematic diagram of the testing assembly. Temperature was determined using digital electronic thermometer type. The assembly was kept isolated from the interference of ambient conditions with the help of a clear transparent cellophane box. The mass flow rate of the distilled water was kept constant at 50, 100 and 200 ml/h. Efficiency of the solar heat collection was calculated in $J.cm^{-2}.s^{-1}$ taking the heat capacity Cp value of water under ambient pressure $4.1819\ J.g^{-1}$. The outer surface area of the glass coils assembly was determined from $A = L_{(tube)}.\ L_{(circumflex.)}$.. L of the tube was computed from the weight of water filling the tube (the specific density value of water at 25°C= 0.998) $Wt_{(water)}$ x 0.9998 x πr^2 (r = outer radius – 2 thickness of the tube)

$\sum$ A= $(Wt_w\ /r+\ 2tw)_1$ + $(Wt_w\ /r2tw)_2$ +…..+ $(Wt_w\ /r+ 2tw)_n$..(1)

The amount of solar heat collection was determined from

$Wt_w.Cp\ .\Delta T/\ A.\ t(s)$ ……………..(2)

Fig.1 shows a schematic diagram of the carbon-cuprous oxide composite assembly used to collect solar heat. It is built up from 5 glass coils connected in series. Temperature of the flowing water was recorded with the help of a sensitive thermometer.

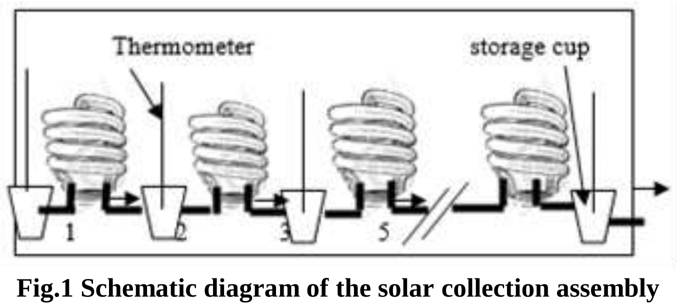

Fig.1 Schematic diagram of the solar collection assembly

III. RESULTS

Fig. 2 shows SEM image of the prepared nanocarbon particles from the fly ash.

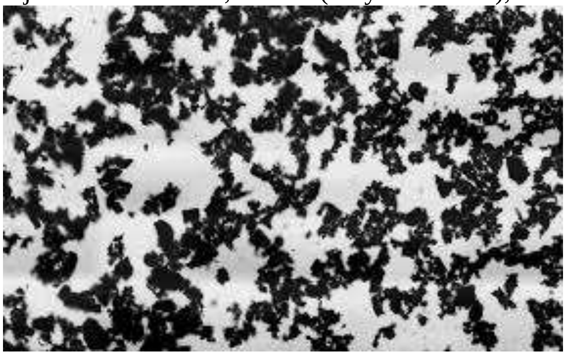

Fig. 2 SEM image of the prepared nanocarbon particles.

Table 1 shows the physical properties of the waste carbon used in this study.

Table 1 The physical properties of the waste carbon

Property	Value	Chemical analysis	
Apparent density g/cm^3	0.75	element	Weight %
Grain size, % wt > 10 um <10 >1 um <1 um	 4 8 88	C Fe Ca Ash	94.72 0.80 0.12 4.36
pH	7.8		100.00

Table 2 shows the working data of the solar collection assembly. The glass tube has 8.925 – 8.85 mm inner diameter, 1 mm wall thickness and the number of circles/ coil is 5 circles.

Fig. 3 shows a FE-SEM photograph of the prepared carbon nanotubes.

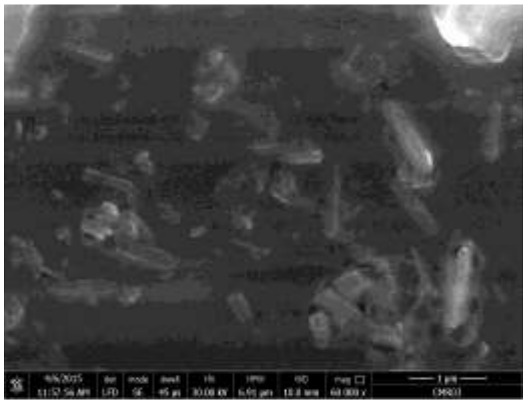

Fig. 3 FE-SEM photograph of the prepared carbon nanotubes.

Table 2 The working data of the solar heat collection assembly

No. of glass coils	Number of glass coils / assembly		
	1	5	5
surface area	248..8 -295	1000 – 1300	1250 -1500
Water mass flow rate, g/min.	10 - 50	10 - 50	10 - 50
T, of water inlet, °C	20	20	20
Weight of C, Cu , ug cm^2	4 0-4	4 0-4	4 0-4
C:Cu Wt ratio	1:5 1:1	1: 5 1:1	1: 5 1:1

Fig. 4 shows the weight of carbon nanoparticles deposited on glass tube surface as affected by time of application
It can be seen that the weight of carbon deposited increases with increase of the carbon concentration in water and number of application as well as concentration of carbon in slurry.

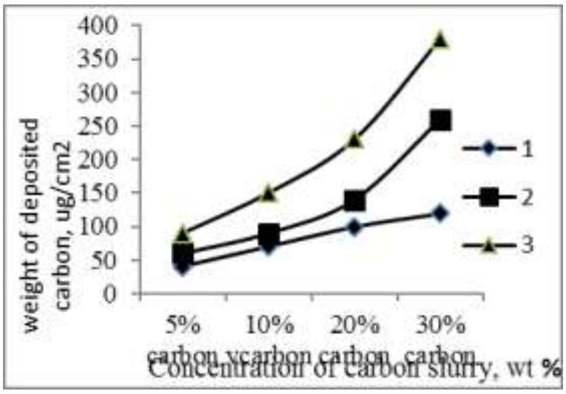

Fig. 4 weight of carbon nanoparticles deposit on glass surface as affected by number of application and concentration of carbon in slurry.

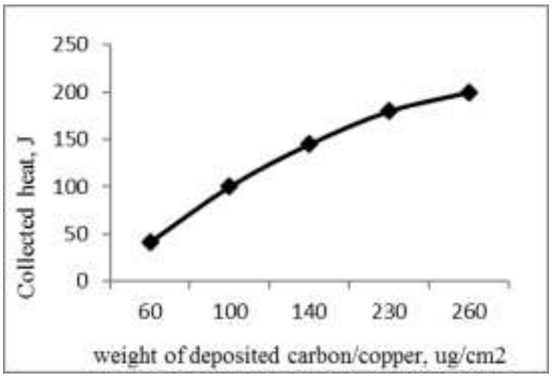

Fig. 5 The effect of copper/carbon weight ration on the extent of the collected solar heat

Fig. 5 shows the effect of copper/carbon weight ratio on the extent of the collected solar heat. It is seen that increasing the copper ratio brings about a corresponding increase in the extent of the collected solar heat.

Fig. 6a and 6b show the XRD of the product of reducing copper nitrate with hydrazine hydrate for 5 minutes. It is seen that Hydrazine hydrate partially reduces the copper salt to cuprous oxide. With more hydrazine dose and with long time of treatment, complete reduction takes place

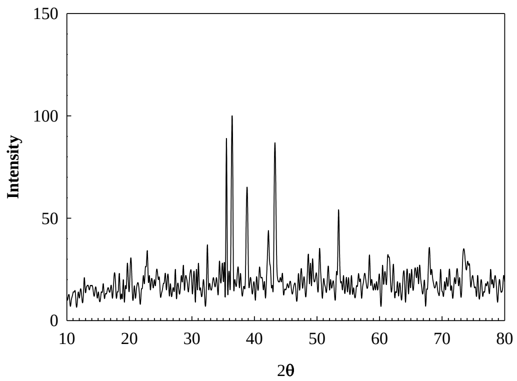

Fig. 6a The XRD pattern of cuprous oxide prepared by reduction of copper nitrate with hydrazine hydrate

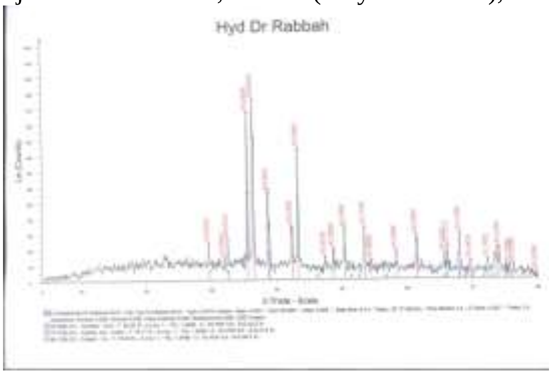

Fig. 6b A photograph of The XRD pattern of cuprous oxide prepared by reduction of copper nitrate with hydrazine hydrate

Fig 7 shows the extent of the collected solar heat as a function of weight of deposited cuprous oxide: carbon on glass surface. It is seen that the extent of the collected heat from sun increases with increase in cuprous oxide: carbon weight ratio. The maximum collected heat energy obtained with 4:6 carbon to cuprous oxide amounts to 380 J and increases to 540 J with 1:4 carbon to cuprous oxide.

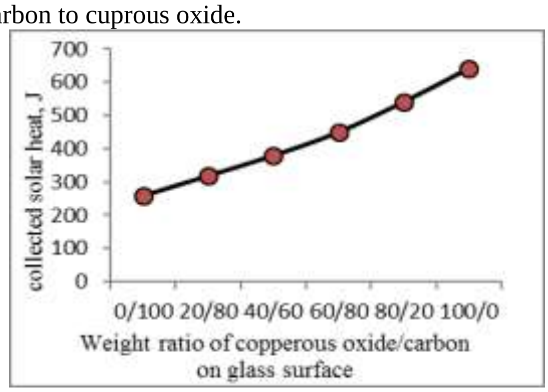

Fig. 7 Effect of weight ratio of cuprous oxide / carbon of the deposited carbon particles (on glass surface) on the extent of the collected solar heat.

Fig. 8 shows the extent of collected solar heat as a function of the weight ratio of cuprous oxide/carbon on the glass surface. Experiments were carried out during March 2015 whereby the average ambient temperature in the day was around 19°C. The testing assembly is built up of 5 glass coils coated with cuprous oxide/carbon layer having different densities of 1.5 ug/cm^2 to 6 ug/cm^2

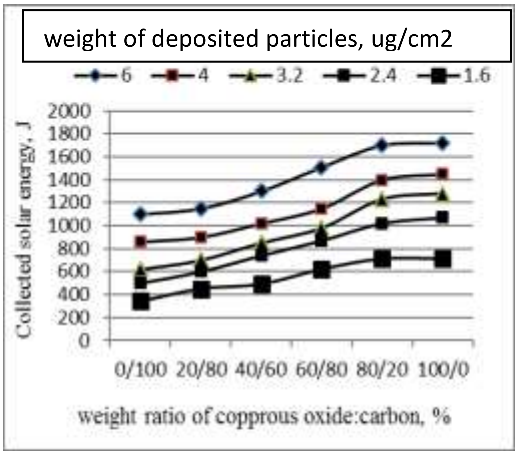

Fig. 8 the extent of collected solar heat as a function of the weight ratio of carbon/cuprous oxide/composite on the glass surface.

It can be seen that the extent of the collected solar heat energy increases with increase in cuprous oxide ratio and the weight of the deposited composite. It is also seen that the maximum heat collected from solar resource amounts to 720 J obtained with coating of 1.6 ug/cm^2. .The extent of the collected heat increases to 1280 and 1720 .J with a coat weight of 3.2 ug/cm^2 and 6 ug/cm^2 respectively.

IV. DISCUSSION

When the solar heat arrives at the surface of the earth, it has been attenuated twice by both the atmosphere (6% by reflection and 16% by absorption and the clouds (20% by reflection and 3% by absorption [16]). Another 51% (89 PW) of the total incoming solar radiation reaches the land and the oceans [17]. It is evident that, despite the attenuation, the total amount of solar heat available on the Earth is still of an enormous amount, but because it is of low-density and intermittency, it needs to be collected and stored efficiently [17]. Kalogirou [18] reviewed several different types of solar thermal collectors that were in common use, and provided relative thermal analyses and practical applications of each type. However, the technologies involved in solar collectors have been much improved since that review was published, so that some of the latest collectors, such as PVT(Photovoltaic-Thermal) collectors, were not available in time for inclusion. Both carbon black powder and nanocuprous oxide had good absorption properties of heat and solar heat in the whole wavelength ranging from 200 to 2,500 nm. Nanocuprous oxide exhibited a shear thinning behavior. The shear viscosity increased with the increasing volume fraction and decreased with the increasing temperature at the same shear rate. The thermal conductivity of carbon black nanocuprous oxide increased with the increase of volume fraction and temperature. Carbon black nanocuprous oxide had good absorption ability of solar heat and can effectively enhance the solar absorption efficiency. Due to its thermal conductivity and coefficient of thermal expansion (CTE) vapour grown carbon nanofibres (VGCNFs) are one of the most promising reinforcing materials of metal matrix composites(MMCs) for the thermal dissipation of future power electronics [19]. VGCNFs provide currently an immediate availability at industrial scale and excellent performance-cost ratio for industrial production of electronic packaging components, specifically for space applications where higher power densities and weight save are crucial. Table 3 shows the thermal conductivity properties of carbon and cuprous oxide.

Table 3 the thermal conductivity value of carbon and cuprous oxide [19]

material	Density, $g.cm^{-3}$	Therm. Conductivity W/m.K
Carbon nanotubes	2	6600
Copper	5.9	150-300

Results given in Figs.3 through 5 comply with the physical properties of carbon nanotubes and cuprous oxide used in this work to collect solar heat. As copper percentage in the composite goes up, the collected heat increases. The composite material receives heat from solar rays whereby thermal conductivity plays its role to convey that heat to the glass wall of the coil. As glass is relatively bad thermal conductor, a thermal gradient is established across the outer and inner surfaces of the glass tube. Heat would then transfer to the flowing water when a temperature potential is set in across it and the surface of the inner glass wall. It is worthy to note that such temperature potential is highest in the first coil and decreases gradually with the next coils. At certain coil, no heat would transfer if the temperature gradient diminished to zero.

Heat transfers from the falling sun rays to the water in the glass tube across two layers; the carbon-cuprous oxide composite in a horizontal/vertical two dimensional mechanism; and to the glass wall of the tube in one dimensional mechanism. Thermal equations for these heat transfer models are reported elsewhere [20]. .The collected heat from the test assembly amounts to 1720 Joule from a total heat transfer surface area of 1500 cm^2 corresponding to 11.46 kJ per meter square. Worthy to note that the test was carried in the beginning of March 2015 in Cairo. Egypt. The collected heat would be further increased when collection takes place during hot summer months whereby the sun rays becomes perpendicular to the earth. The tested tube assembly is the absorber of the solar water heater. Traditional solar heat collectors are built up of double tubes. The inner tubes absorb solar energy converting it into heat for use in water heating. The tube is transparent allowing light rays to pass through with minimal reflection. The inner tube is coated with a special selective coating (Al-N/Al) which features excellent solar radiation absorption and minimal reflection properties. The top of the two tubes are fused together and the air contained in the space between the two layers of glass is pumped out while exposing the tube to high temperatures. This "evacuation" of the gasses forms a vacuum, which is an important factor in the performance of the evacuated tubes.

V. CONCLUSION

The output conclusion of this study is as follows.

1. The heat collection assembly has proven suitable to heat water for domestic use.
2. The carbon/cuprous oxide composite is found good heat collector from the falling rays of sun
3. The appropriate composite composition of nanoparticles of carbon/cuprous oxide amounts to 1:1.
4. Glass coils of spent saving energy lamps or other sources are found convenient to construct a heat collector assembly.

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