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Effect of Nano-CuO grain size on Heat Transfer Performance of Copper Substrate

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Abstract:

This paper presents the experimental results of nano copper oxide coating on the heat transfer performance of the copper substrate. The thermal effect of Nano copper oxide coating investigated by using an experimental thermal system consisted of three sections: evaporator, adiabatic, and condenser. The copper oxide (CuO) coating performed for the inner surface of copper pipe using an electrochemical oxidation method in an oxalic acid bath with applying a voltage of 5 volts at ambient temperature. The experimental tests performed for coated and uncoated copper pipes. The effects of CuO grain size on specific surface area (SSA) and material characteristics feature are investigated. Characterization of the CuO coat-ing were conducted using X-ray diffraction (XRD) and Field Emission Scanning Electron Microscopy (FESEM). The aver-age grain size of the oxide coating was ~44 nm, with an average coating thickness of ~11 micrometers. The CuO coated substrate showed lower thermal resistance property compared to uncoated substrate. However, the SSA of the coated substrate in decreased with the increasing of CuO grain size. Results show that the Nano-copper oxide coating enhances the heat transfer performance of the copper pipe, as a result of the increase in the specific surface area of Nano range grain size CuO by reducing the thermal resistance, and increasing the thermal conductivity. This result opens a new avenue to overcome the industrial challenge in the development of the heat exchangers design with Nano-coating with increased heat transfer efficiency and reduced size of the heat transfer equipment.

Keywords: *Copper; CuO; Electrochemical oxide Coating; Thermal effect; Heat Transfer performance.*

I.INTRODUCTION

Copperalloys are commonly used in the manufacturing of heat transfer equipment like heat exchanger boiler and cooling towers, because of their good heat transfer and corrosion resistance properties. Copper's thermal conductivity is higher than aluminum and stainless steel [1]. It has suitable corrosion resistance to be used in untreated acidic water environments, whereas aluminum can releases hydrogen gas in this environment, which may cause industrial hazardous [2]. Copper also has high anti-microbial properties, as compared with stainless steel, which gives it the advantage to use it in the food industry [3]. Therefore, copper alloys become the primary manufacturing material choice of safe, efficient heat transfer equipment.

Using copper in contact with industrial water usually causes a serious industrial problems. Copper contamination in one of these industrial problems that occurs in boiler feed water system as a results to the corrosion of copper boiler’s tubes in the feed water environment. Copper deposits on boiler tube surface can depress the melting point of the boiler tube material, causing early failure known as liquid metal embrittlement [4], [5]. In this research copper oxide coating used as a corrosion resistance coating to prevent such industrial problem. The coating was conducted using anodizing technique in oxalic acid solution ($H_2C_2O_4$) of concentrations between 0.1 - 0.5 M, at temperature range between 0 – 24 ° C and applied voltage between 7.5 - 9 Volts. The grain size of the anodized copper coating is an essential characteristic because of its relevance affecting in the surface area. It represents the size of the partitions of surface material particles. Reducing surface grain sizes lead to separate materials surface particles into smaller partitions and different shapes, which increase the total surface area. Therefore, more surface area can be achieved by reducing surface grain size.

From previous results [6] it was observed that the characteristic properties of copper was enhanced at the Nano scale grain size of electrochemical oxide copper coating, by enhancing the surface to volume ratio and increasing the quantum effect that controls material properties.

The electrochemical oxide (ECO) also known as anodizing process it is an electrochemical surface treatment used to converts metal surface into hard, corrosion-resistant oxide coating; this coating can be used in various applications as shown in Fig. 1 [7], [8]. According to the mechanical, electrical, and physical properties of the anodized coating, it can contribute to a variety of scientific applications like gas detection sensors [9], super hydrophobic surfaces, high-temperature superconductors [10] and solar cells [11].In 2014 researcher [12] reviewed the characteristics of the copper oxide formed by electrochemical anodization in caustic soda solution at 90 ° C. This study discussed the effects of solution concentration, temperature and current density on the coating characteristics. It was reported, in this study, that the chemical, optical, thermal properties can be enhanced by controlling operating parameters of the anodized coating.

The grain size of the electrochemical oxide coating is the most effective parameter on various properties. Controlling the Nano scale grain sizes and its microstructures are considered as a successful method to develop new mechanical, physical and chemical properties for the coated surface to serve a specific scientific requirement [7], [13], 14]. Therefore, the main aim of this research is to study the effect of Nano copper oxide coating on the heat transfer performance of the copper substrate.

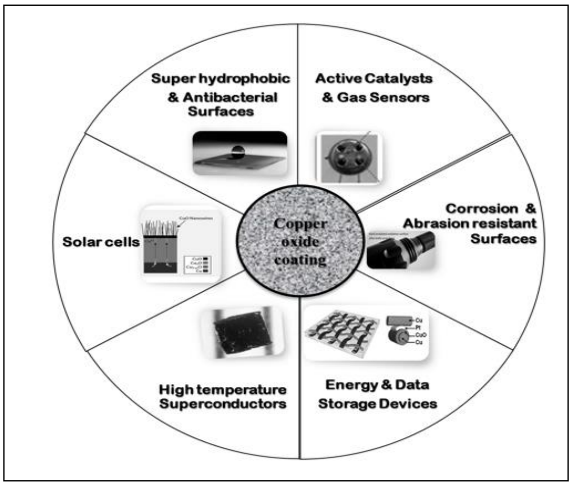


Fig. 1.Application of electrochemical oxide process or anodizing process

II.EXPEREMENTS

The electrochemical oxide (ECO) coating was performed on a cop-per substrate using anodization method in 0.1M oxalate containing solution at ambient temperature and 5 Voltage [14]. Before anodization, the copper sample surface was chemically cleaned in an acid bath and washed in acetone, distilled water and then dried in air at room temperature. Figure 2 shows an illustration of the experimental set-up of electrochemical oxide coating. It consisted of; (a) electrolyte container, (b) counter electrode, (c) reference electrode, (d) working electrode, (e) holder, (f) magnetic stirrer, (g) heater mantel, (h) circulating cooling water jacket, (i) inlet of cooling water, (j) outlet of cooling water, (k) temperature indicator of cooling water, (l) temperature indicator of solution, (m) potentiostat. The experiments were performed in AutolabPotentiostat/Galvanostat PGSTAT AUT86037 supported with (NOVA 1.10 Software) using linear sweep voltammetry (LSV) method at scan rate 0.02 V/Sec. The electrochemical cell was consisted of three electrodes, Platinum foil as counter electrode, copper as working electrode and silver/silver chloride Ag/AgCl as reference electrode.

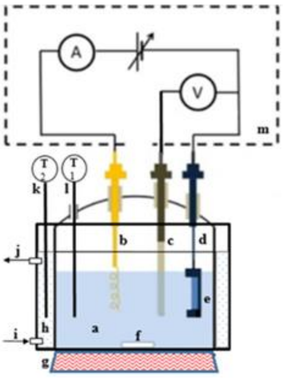


Fig. 2. The experimental set-up of electrochemical oxide coating process.

III. RESULT AND DISCUSSION

The electrochemical oxide coating characteristics was investigated by using X-ray diffraction (XRD) and Field Emission Scanning Electron Microscopy (FESEM). The coated samples were scanned through XRD in the range between 20-80°. The results of the X-ray diffraction pattern in Fig. 3 showed the prominent peaks which are compatible with the diffraction pattern of the cupric oxide Standards Diffraction card, JCPDS 05-0661, the peaks were at 38 and 65° referring to (111) and (022) planes, as represented by two obvious peaks at 38.7° and 43°.

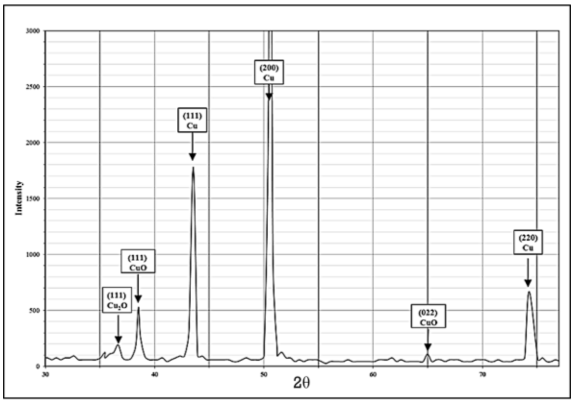
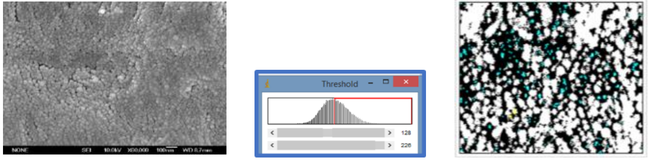


Fig. 3.X-ray diffraction profile of anodized coating

The morphology and microstructure of coated surface in terms of grain size and porosity were investigated using FESEM with image analyzer software. The grain size was measured by linear intercept analyzing method, ASTM E112 - 13(2015). Analysis was made on the sample surface in four straight lines with total length (L) of various directions (0°, 45°, 90°, 135°). The Pores percentage area of the samples were measured according to ASTM E2109 - 01(2014) Standard Test Methods. The percentage of pores on a selected surface area was measured using image analyzer software. Small pores of less than one pixels, were excluded and considered as noise, as shown in Figure 4.



a **b** **c**

Fig. 4. The microstructure of coated surface a) original FESEM image, b) gray threshold analyzing, d) resulted analyzed image

The average grain size of the anodized copper coating was ~44 nm, with an average thickness of ~11 micrometers and the average pores percentage area of the coatingwas ~8.5 % as shown in figure . From Fig 5 it can be seen clearly the morphology of Nano CuO coated substrate in oxalate solution.

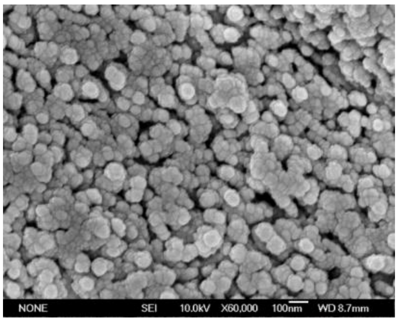


Fig. 5.FESEM image at 60,000x magnification of anodized copper coating prepared in oxalate solution

To investigate the effects of grain size for the Nano CuO coating on heat transfer performance, a thermal experimental system was modeled, as shown in Figure 6.The experimental system consists of high purity copper pipes 99.99%, with 3.18 mm outside diameter, 1.55 mm inside diameter, and 0.81 mm wall thickness. The pipe was bent into six paths passing through three sections, evaporation, adiabatic and condensation. The evaporation section consisted of controllable electrical heater mantel, while the condensation section consisted of circulating cooling water bath. Skin temperatures of the pipe was measured by fixing eighteenth temperature indicators in the six paths of the bent pipe, as illustrated in Figure 6.

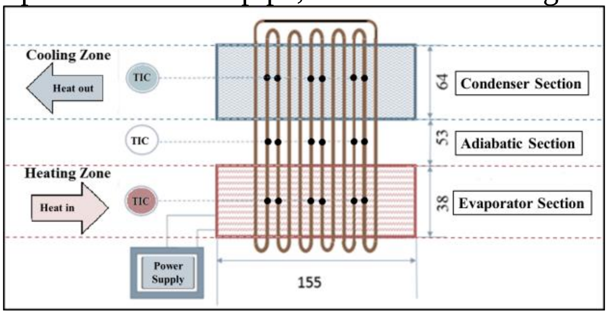


Fig. 6. Experimental Thermal System

In the evaporator sectionthecopper pipes were heated up using a heater mantle controlled with power supply. The heating started at the cooling zone steady-state temperature of 20 °C. All the readings of heating loads and temperatures difference in the evaporation and condenser sections were recorded. For a more accurate result, the average of temperature differences, for each of the six paths, were taken. Thermal experimental tests for the coated and un-coated copper samples were performed. The thermal resistance was measured according to the following equation [15].

$$R= (\Delta T) /Q_h \qquad (1)$$

Where;

ΔT; is the temperatures difference

Q_h; is the input heating power

The experimentalresults showed that the thermal resistance for the coated copper samples was lower than that of the uncoated samples, as shown in Figure 7. This result indicates the enhancement of the heat transfer performance for the anodized coated sample as compared with the uncoated sample. That enhancing is due to the Nanostructure of the coating's grains, which can improve the properties of the material surface by increasing the surface to volume ratio. In the Nanostructure, coating particles get smaller, and the surface area to volume ratio gets larger. Hence quantum effect improved.From the same figure, it was also observed that thethermal resistance was decreasing with the increasing of the heating load from 25 to 250 Watt.

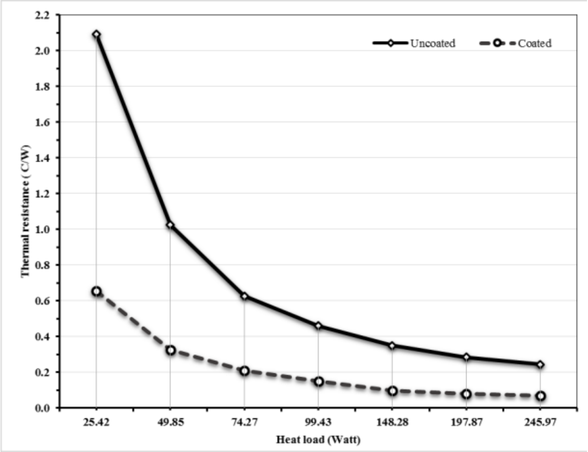


Fig. 7. Thermal resistance as a function of heating loads for coated and un-coated samples

The importance of grain size as an essential material characteristic appeared from its effects on the surface area whereby reduced grain size increased surface area as a result of the partitions of surface material particles into smaller parts leading to rising in the total surface area. Increase surface area results in enhancing the surface to volume ratio and increasing the quantum effect which controls material properties [6]. Therefore, nanotechnology contributed to the enhancement and development of material mechanical properties, corrosion resistance, and heat transfer performance of substrate material.

The specific surface area (SSA) was measured from the material surface grain size according to the following relation:

$$SSA=6/ (\rho \times d \quad) \; (2)$$

Where,

ρ is the material density

d is the grain size diameter

Results showed that the surface area is increased dramatically with the decreasing of Nano coated surface of grain size leading to the better heat transfer performance of substrate as shown in Figure 8.

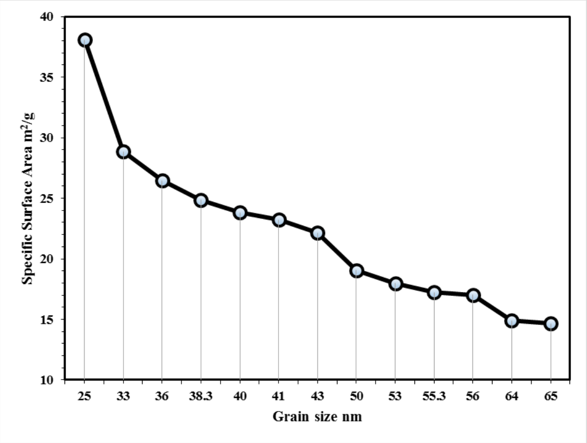


Fig. 8. Specific Surface Area of coated surface as a function of coating's grain size

For more investigation on the thermal effect of the anodized coating a thermal investigation for heat transferred through coated sample was made to evaluate the effective heat transfer capability of the samples, effective thermal conductivity at the specific heating flux was measured using the following equations;

$$K_{eff}= (Q \times L_{eff})/(\Delta T \times A_c) \qquad (3)$$

$$L_{eff} = (L_e/ 2) +L_a+ (L_c/2) \qquad (4)$$

Where;
Keff; is the effective thermal conductivity W/m.k
Ac; is the total cross-section area of the tested samples cm^2
Leff; is the total effective length
(Le, La, Lc); are the lengths of the evaporation, adiabatic, and condensation sections of copper samples.

$$q = ((K_{eff})/ (L_{eff}))/((\Delta T\times A_c)) \qquad (5)$$

$$\text{Heat flux } q =Q/A_c \qquad (6)$$

Where;
q; The heat flux, or the rate of heat transfer per unit area, (W/m^2)
Q; transferred heat joules (J)

Results show the increase of the effective thermal conductivity with the raises of the specific surface area of the sample, as shown in the Figure 9.

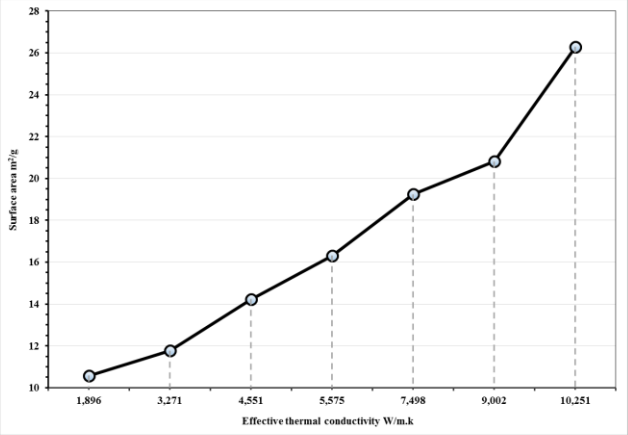


Fig. 9. Effective thermal conductivity (W/m.K) of coated samples as a function of Specific Surface Area

The previous resultshowed that the effective thermal conductivity is increased more than 5 times with the increase of specific surface area 9.6 m^2/g (from 10.6 to 26.3).

IV. CONCLUSION

The following notes was concluded from previous results;

1. The electrochemical oxide copper coating was successfully fabricated on a copper substrate in oxalate containing solution. The average grain size of the anodized copper coating was ~44 nm, with an average thickness of ~11 micrometers and the coating pores percentage area was ~8.5 %.
2. Thermal testes for coated samples shows that thermal resistance was decreased after electrochemical oxide process and with the increasing of the input heating load.
3. The heat transfer of electrochemical oxide coated sample was enhanced by increasing the specific surface area by 2.5 times with the increase of average coatings grain sizes of 40 nm.
4. Increasing of surface area improves the heat transfer by increasing the effective thermal conductivity by 5 times with the increases of specific surface area by 10 m^2/g.

This investigation can facilitate many industrial challenges to develop the design of heat exchangers equipment by increasing the heat transfer efficiency and reducing the active surface area using Nano-coating.

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