



THERMAL PROPERTY OF CARBON RESIN ELECTRODES DEVELOPED FOR ELECTROCHEMICAL TREATMENT OF WATER AND WASTEWATERS

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ABSTRACT

In this paper, thermal conductivity of carbon resin electrodes developed for electrochemical treatment of water and wastewater was investigated. Carbon resin electrodes were developed from used dry cells and resin using non-heat treatment processes. Thermal conductivity of the electrodes was measured and effects of particle size, compacting pressure, carbonisation temperature and percentage of the resin used on thermal conductivity of the electrodes were monitored using standard method through RE 890G and ALDA AVD 890G thermocouples at two different points (5cm apart). Effective thermal conductivity of the electrodes was modelled using Okazaki et al model.

The study revealed that thermal conductivity of carbon resin electrodes ranged from 1.39 W/K. cm to 2.24 W/K. cm. Thermal conductivity of the material increased with decreased particle size (1.48 to 2.24 W/K. cm with 245 to 45m) and decreased percentage resin (1.49 to 2.20 W/K. cm with 12 % to 1%), decreased with decreased compacting pressure (2.12 to 1.44 W/K. cm with 110 MN/m² to 60 MN/m²) and carbonization temperature (2.12 to 1.39 W/K. cm with 260 °C to 30° C). Okazaki et al model agreed reasonably with the experimental data with correlation coefficient of 0.3206 and coefficient of determination 0.9917. It was concluded that particle size, compacting pressure, carbonization temperature and percentage resin play important role in thermal conductivity of carbon resin electrodes.

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1.0 INTRODUCTION

Electrochemical treatment process is an important water and wastewater treatment method in water resources management and environmental pollution control. The main

components of this treatment process are electrodes, wastewater and direct current source. The importance of electrodes in electrochemical treatment process has been well documented (Chen, 2004, Oke *et al.*, 2017). In addition, carbon electrodes are

known to be important during this treatment process due to their resistance to chemical attack (Oke, 2006). In previous studies (Oke *et al.*, 2007a and b, 2010 a and b), temperature was identified as a factor that can influence efficacy of the treatment process and stability of carbon resin electrodes (Oke 2009a and b), which indicates that thermal conductivity plays an important role in electrochemical treatment process. Energy transfer arising from the temperature difference between the adjacent parts of the body is known as heat conduction. It is well known that higher temperatures increase the energy of the electrons and permit heat to be transferred by lattice vibration. In metal, the thermal conductivity often initially decreases with temperature, later becomes nearly constant, and then increases slightly, as in iron (Adem and Hakan, 2007). Conductivity of some materials decreases continuously when some materials are heated (e.g. when aluminum is heated) but increases continuously (when platinum is heated, Oke *et al.*, 2007a and b). Thermal conductivities of metals have effects on several properties. Harden-ability and weld-ability of steel are affected by their thermal conductivity. In previous studies, (Oke *et al.*, 2007a, b and c) significant factors for the stability of carbon resin electrodes are particle size, percentage of resin used and compacting pressure. These indicate that the determiners of the carbon resin application include thermal conductivity level. Thermal conductivity (k) is a measure of the rate at which heat is transferred through a material. The conductivity relates the heat (Q) transferred across a given plane of area (A) per second when the temperature gradient exists (Adem and Hakan, 2007, Nelkon, 2002, Abou

– Sena *et al.*, 2007). The mathematical expression is as follows:

$$\frac{dQ}{dt A} = -k \frac{\Delta T}{\Delta x} = q \quad (1)$$

where; Q is the heat energy transferred per unit time per (J/s; W), A is the cross-sectional area (m²), k, is the thermal conductivity, Δx is the change in the longitudinal direction (m) and ΔT is the change in the temperature (°C).

The amount of heat to be transferred through anybody depends on a number of factors such as the particle shape, porosity, temperature range, solid constituents, moisture contents, pressure etc. (Wu *et al.*, 2007, Sighn *et al.*, 2007). More on thermal conductivity of carbon and graphite electrodes can be found in Lin *et al.*, (1993), Zhu *et al.*, (2007); Yuen *et al.*, (2007); Zhang *et al.*, (2008); Yang *et al.*, (2009); Zhou *et al.*, (2010); Ramvir and Sharma (2011), Oke *et al.*, (2012); Kuzhir *et al.*, (2013); Rahmani *et al.*, (2014) Arohi *et al.*, (2015), Yue *et al.*, (2017); Leemsuthep *et al.*, (2017); Ravi *et al.*, (2017), Bera *et al.*, (2018); Eungjelee *et al.*, (2018), Chang and Dinh (2019), Mohammed and Norbert (2019). Literature stated clearly the relationship between materials and their thermal conductivity and detailed importance of carbon electrodes, but literature on thermal conductivity of carbon resin electrodes and factors that influence thermal conductivity are rare. This identification calls for studies on thermal conductivity of carbon resin electrodes and factors that influence its performance in electrochemical treatment of water and wastewaters. The main objective of this study therefore is to examine effects of selected factors (temperature, particle size of carbon, compacting pressure and percentage

of resin) on thermal conductivity of carbon resin electrodes developed for electrochemical treatment of water and wastewater in a laboratory scale.

2.0 MATERIALS AND METHOD

Discarded dry cells (size D R20 UM-1) were collected in Nigeria (Figure 1a). These dry cells were sectioned; graphite (carbon) were removed and pulverised (Figure 1b). Powdered graphite was sieved into different particle sizes (Figure 1c). A known mass (10 grams) of the powdered graphite was mixed

with an organic binder, moulded into 2.5-centimetre diameter, a 10-centimetre-long electrode using locally fabricated extruder and plunger and a hydraulic compacting machine (Figure 1d). Longitudinal temperatures change of developed carbon electrodes were determined using RE 890G and ALDA AVD 890G thermocouples at two different points (5 cm apart). Revotherm hot plate (manufactured by Rodwell Science Instrument England) was used as source of heat temperature. Effects of graphite particle size, compacting pressure, carbonization temperature of the electrodes and percentage resin on thermal conductivity



Figure 1a: some of the collected dry cells

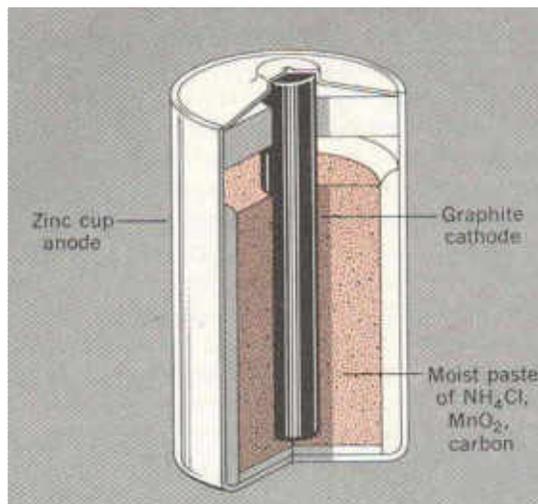


Figure 1b: sectioned dry cell



Figure 1c: Some collected Carbon rods/graphite



Figure 1d: Powdered graphite

of the electrode were studied. A known mass (10 grams) of the electrode was soaked in 300 ml of distilled water for 24 hours (for wetness to a saturation level). The mass after saturation was determined and porosity of the electrode was computed for using equation (3).

Effects of carbonization on porosity of the developed electrodes were conducted and used in the modelling effective thermal conductivity (which was computed and modelled using Okazaki et al model). The model was selected based on the shape of the

electrodes (spherical).

Where; V_v is the volume of the void, W_2 is the weight of saturated the electrode (g) and W_1 is the dry weight of the electrode(g).

$$V_v = W_2 - W_1 \quad (2)$$

Where; V_v is the volume of the void, W_2 is the weight of saturated the electrode (g) and W_1 is the dry weight of the electrode(g).

$$\varepsilon = 100 \left(\frac{V_v}{2\pi r h} \right) \quad (3)$$



Figure 1e: Some of the carbon resin electrodes developed



Figure 1f: some of the carbon resin electrodes used electrodes



Figure 1g: Crusher (used to break the rods)



Figure 1h: Grinder (Used to reduce particle size of the rods)

Where; r is the radius of the electrode (2.5×10^{-2} m) and h is the length of the electrode (10.0×10^{-2} m)

Models of effective thermal conductivity of carbon resin electrodes:

Several models have been presented on thermal conductivity to predict the effective

thermal conductivity of a given material beds (Lin *et al.*, (1993), Abou –Sena *et al.*, (2007), Wu *et al.*, (2007), Wang *et al.*, (2007), Ramvir and Sharma, (2011), Arohi *et al.*, (2015), Ravi *et al.*, (2017), EungjeLee, *et al.*, (2018), Chang and Dinh (2019), Mohammed and Norbert (2019) . Some of the models available are as follows:

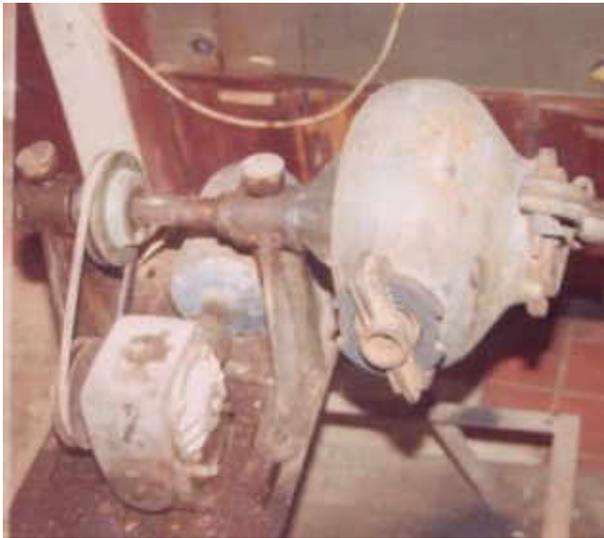


Figure 1i: Power ball roller (used to powder the rods)



Figure 1j: Plunger (used for moulding the electrode)

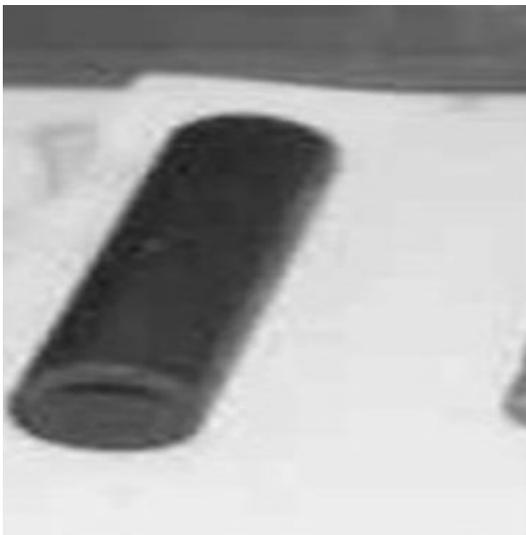


Figure 1k: Extruder (used for moulding the electrode)

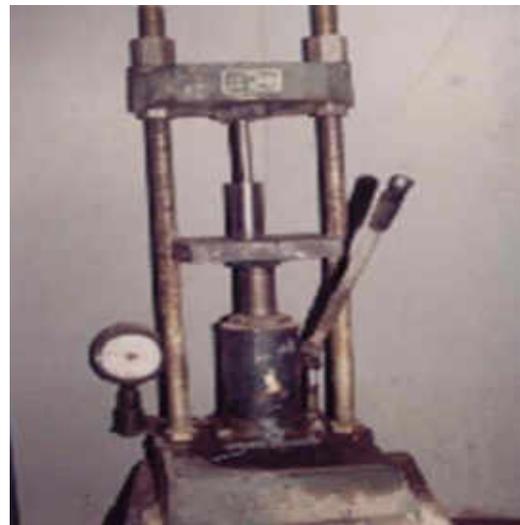


Figure 1l: Hydraulic compressive machine (used in moulding the electrode)

a. Schlunder, Zehner, and Bauer model

The Schlunder, Zehner, and Bauer (SZB) model is widely used to predict the effective thermal conductivity of material beds (Abou –Sena *et al.*, 2007). This model can be used for beds of materials of any shape and size

distribution, but having the same thermal conductivity. The SZB model takes into account the conduction and radiation effects throughout the bed. Based on parallel resistances, the bed effective thermal conductivity, k_{eff} , can be calculated by using the following equations (Abou –Sena *et al.*, 2007):

$$\frac{k_{eff}}{k_g} = (1 - (\sqrt{1 - \epsilon})) \left[\frac{\epsilon}{(\epsilon - 1) + \frac{k_g}{k_D}} + \frac{\epsilon k_R}{k_g} \right] + \sqrt{1 - \epsilon} \left[\phi \frac{k_s}{k_g} + (1 - \phi) \frac{k_{so}}{k_g} \right] \tag{4}$$

Where; k_R is the equivalent thermal conductivity due to radiation, k_s is the thermal conductivity of material, k_{eff} is the effective thermal conductivity of a material packed bed, k_D is the equivalent thermal conductivity due

to molecular heat flow, k_g is the gas thermal conductivity at atmospheric pressure, and ϕ is the material-to-material contact area,

$$\frac{k_{so}}{k_g} = \frac{2}{a} \left[\frac{\left(B \frac{k_s}{k_g} + \frac{k_R}{k_g} - 1 \right) \frac{k_g}{k_D}}{a^2 \frac{k_s}{k_g}} \right] \ln \left[\frac{\left(\frac{k_s}{k_g} + \frac{k_R}{k_g} \right) \frac{k_g}{k_D}}{\left[B \left(1 + \left(\frac{k_g}{k_D} - 1 \right) \left(\frac{k_s}{k_g} + \frac{k_R}{k_g} \right) \right) \right]} \right] - \left(\frac{B - 1}{a} \right) \frac{k_g}{k_D} + \frac{B + 1}{2B} \left[\frac{k_g}{k_D} - B \left(1 + \left(\frac{k_g}{k_D} - 1 \right) \frac{k_R}{k_g} \right) \right] \tag{5}$$

$$a = \left[1 + \left(\frac{k_R}{k_g} - B \frac{k_D}{k_g} \right) \frac{k_g}{k_s} \frac{k_g}{k_D} - B \left(\frac{k_g}{k_D} - 1 \right) \left(1 + \frac{k_R}{k_s} \frac{k_g}{k_s} \right) \right] \tag{6}$$

The deformation factor, B can be calculated as follows:

$$B = C \left[\frac{1 - \epsilon}{\epsilon} \right]^m \tag{7}$$

Both C and m should be determined from the experimental data. The amount of heat transfer by radiation depends on the absolute temperature, T , of the packing, the radiation characteristics of the materials, and the

packing geometry. For small temperature gradients, the radiative heat transfer is described by Damkohler's equivalent thermal conductivity, k_R , as follows (Abou –Sena *et al.*, 2007):

$$\frac{k_R}{k_D} = \frac{0.04 \delta}{\left(\frac{2}{\gamma} - 1 \right)} \left(\frac{T}{100} \right)^3 \frac{X_R}{k_g} \tag{8}$$

σ is the Stefan–Boltzmann constant and γ is the radiative emissivity. X_R is the effective

radiation length that characterizes the distance between the materials' surfaces

$$\frac{k_g}{k_D} = 1 + \frac{2\lambda}{X_D} \left[\frac{2}{\alpha} - 1 \right] \tag{9}$$

$$X_R = R_f d \text{ and } X_D = D_f d \tag{10}$$

Where; λ is the mean free path at atmospheric pressure, R_f is the shape factor for the interstitial energy transport by radiation, D_f is the shape factor for the interstitial energy transport by molecular flow, d is the material diameter, and α is the accommodation coefficient,

thermal conductivity of ceramic powder packed beds. In the model, the material bed was assumed to be composed of identical materials that contact each other over area of πa^2 . The model was simplified by assuming the materials to be cylinders of radius d and height L , aligned in the same x -direction and contacting each other through a contact point of thickness δ and circular area of radius a . The effective thermal conductivity, k_{eff} , of a material packed bed was defined as follows:

b. Shapiro *et al.* model

Shapiro *et al.* (Abou –Sena *et al.*, 2007) published their model for predicting effective

$$k_{eff} = \frac{4}{\pi} k_s \left(\frac{1 + \frac{\delta}{L}}{1 + \frac{\delta}{L} \left(\frac{k_s}{k_g} \left(\frac{1}{1 - \left(\frac{\alpha}{d}\right)^2} \right) \left(\frac{k_\delta}{k_a} + \frac{1}{\left(\frac{d}{\alpha}\right)^2 - 1} \right) \right)} \right) \tag{11}$$

Where, δ is the gap between materials, k_s is the thermal conductivity of solid material, and k_a is the thermal conductivity of solid material within contact region, v is the specific heat ratio of the gas, λ is the mean free path at atmospheric pressure, P_r is the Prandtl number, α is the accommodation coefficient, k_g

is the gas thermal conductivity at atmospheric pressure, p is the gas pressure, and p_0 is the atmospheric pressure. The values of gap thickness, δ , and contact area size, a , are selected to obtain the best possible fit with the experimental data.

$$k_\delta = k_g \left(\frac{B}{\delta p} + 1 \right)^{-1} \tag{12}$$

$$B = \left(\frac{4v}{v+1} \right) \left(\frac{2-\alpha}{\alpha} \right) \lambda \rho P_r^{-1} \tag{13}$$

c. Okazaki *et al.* model

Okazaki *et al.* (Abou – Sena *et al.*, 2007) predicts the effective thermal conductivity of material beds. In this model, the variations in material size and material thermal conductivity were considered. In the model, the heat flux through the material bed is divided into heat flux through the gaseous phase with a relative surface (A_v) and heat flux through the solid phase with a relative surface (A_s). The heat flux is determined by the contributions of the heat transfer through the gas near the contact point and the heat transfer through the material. For material beds formed by spheres that have similar size, the relation between the coordination number, N , and the porosity, ε , was given as follows:

$$N = 13.84 - \sqrt{232\varepsilon - 57.18} \tag{14}$$

Then the effective number of contact points ($n = N/6$) on a hemisphere can be calculated. This unit cell model consists of a solid part and a macro-void part. The fractional areas of the solid part and the macro-void part were calculated as follows (Abou–Sena *et al.*, 2007):

$$A_s = \frac{3(1-\varepsilon)}{2} \tag{15}$$

$$A_v = \frac{3\varepsilon - 1}{2} \text{ for } \varepsilon \geq 0.33 \tag{16}$$

The effective thermal conductivity of the solid part, k_{eff} can be calculated by

$$\frac{k_{eff}}{k_g} = 2n \left[\frac{k}{k-1} \right]^2 \left[\ln[k - (k-1)\cos\theta] - \left(\frac{k-1}{k} \right) (1 - \cos\theta) \right] \tag{17}$$

$$k = \frac{k_s}{k_g} \text{ and } \theta = \text{Sin}^{-1} \sqrt{\frac{1}{n}} \tag{18}$$

The effective thermal conductivity, k_{eff} , is then formulated as follows:

$$\frac{k_{eff}}{k_g} = A_v k_s + A_s k_s \frac{k_{es}}{k_g} \tag{19}$$

More on thermal conductivity of carbon and graphite electrodes can be found in Lin *et al.*, (1993), Ramvir and Sharma (2011), Arohi *et al.*, (2015), Ravi *et al.*, (2017), EungjeLee, *et al.*, (2018), Chang and Dinh (2019), Mohammed and Norbert (2019).

3.0 RESULTS AND DISCUSSION

Figure 2 shows thermal conductivity profiles for each compacting pressure from 60 to 110 MN/ m². For 60 MN/ m² the profile shows a decrease in thermal conductivity over an

increase in percentage resin, which indicates that at higher percentage of resin reductions in thermal conductivity take place. This may be due to insulating property of the binder (resin, an organic compound). The profiles were similar from 60 to 110 MN/ m². However, at a fixed percentage resin (2%), thermal conductivity increased from 1.72 W/ K. cm to 1.98 W/ K. cm when compacting pressure increases from 60 to 110 MN/ m² at 2 % resin. Figure 3 presents thermal conductivity profiles for each compacting pressure from 60 to 110 MN/ m². Each experimental profile is

then compared with the particle size of carbon. For 60 MN/m² the profile indicates a decrease in thermal conductivity over an increase in particle size, which can be interpreted as higher particle size led to reduction in thermal conductivity. This may be due to lower contact surface area in larger particle size and higher contact surface area for fine particles. The profiles were similar for all particle sizes from 45 m to 245 m. However, at a fixed particle size (45m), thermal conductivity increases from 1.92 W/K. cm to 2.24 W/ K. cm when compacting pressure increases from 60 to 110 MN/ m² at 1 % resin. Figure 4 is a plot of thermal conductivity profiles for each carbon particle size from 45 m to 245 m at various heating temperatures. Figure 5 presents thermal conductivity profiles for each percentage resin with compacting pressure from 60 to 110 MN/ m². From Figure 4 each of the experimental profiles was compared with the carbonisation temperature of carbon resin. For temperature of 30 ° C the plot shows an increase in thermal conductivity over an increase in carbonisation temperature, which can be attributed to reductions in percentage of the binder present (insulator) and increased mobility of electrons in the carbon atom. The plots were similar for all carbon particle size (from 45 m to 245 m), but at a fixed temperature (100°C), thermal conductivity decreased with increased carbon particle size from 1.53 W/ K. cm to 1.84 W/ K. cm when carbon particle increases from 45 m to 245 m at 1 % resin. From figure 4, percentage resin of 0.5% resulted in thermal conductivity decreases over an increase in compacting pressure from 60 to 110 MN/ m². These increments in thermal conductivity can be attributed to reduction in pore and insulating

property of the binder (resin). The profiles are similar for all resin percentages (0.5 to 8%) and for compacting pressure (from 60 to 110 MN/ m²). However, at a fixed particle size (45m) and compacting pressure (90 MN/ m², thermal conductivity increased with decreased percentage resin from 1.69 W/K. cm to 1.85 W/ K. cm when percentage resin decreased from 8.0% to 0.5% resin. Figure 6 (a and b) shows the measured thermal conductivity (experimental data) of the carbon resin electrode compared with the prediction of Okazaki *et al.* model in relation to carbonization. From the figure it can be seen that there is an exponential relationship between carbonization temperature and thermal conductivity. The goodness of a model was evaluated by using the coefficient of determination (R²). It is well known that the closer the value of R² to 1 the better is the correlation and relationship. The coefficient of determination of 0.9331 and 0.9588 for experimental data and Okazaki *et al* model respectively as relationship between carbonization temperature and thermal conductivity suggest that exponential relation can explain 93.31% and 95.88 % of the thermal conductivity. Although, coefficient of determination of Okazaki *et al* model, was lower than the measured values the model agrees with experimental data. Coefficient of determination of 0.9917 with exponential equation of $y = 0.3206 \exp^{1.1418x}$ (Figure 6 b) indicates that correlation between Okazaki *et al* model and experimental data is in a reasonable agreement. This result indicates that Okazaki model can be used to predict thermal conductivity of carbon resin electrode. It can be described that thermal property of carbon resin electrodes in relation

to temperature is similar to property of suspended graphite nano particle described in literature (Abou-Sena *et al.*, 2007), thermal Conductivities of Powder-Filled Epoxy

Resins (Lin *et al.*, 1993), enhanced Thermal Conductivity Of Epoxy Arohi *et al.*, 2015) and cobalt oxide electrodes (EungjeLee *et al.*, 2018).

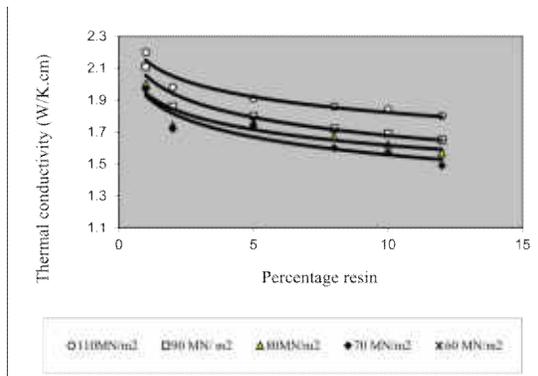


Figure 2: Relationship between thermal conductivity and percentage resin at various compacting pressure, 30°C and 45m particle size

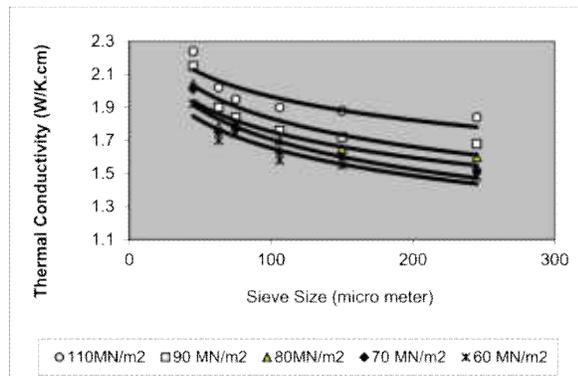


Figure 3: Relationship between particle size and thermal conductivity at various compacting pressure, 30°C and 1 % resin

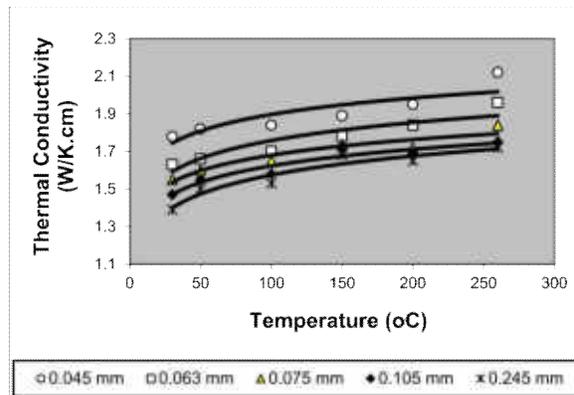


Figure 4: Relationship between thermal conductivity and heating temperature at various particle size, compacting pressure of 100MN/m² and 1 % resin

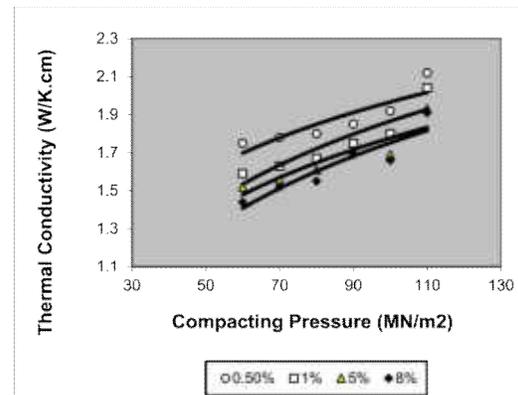


Figure 5: Relationship between compacting pressure and thermal conductivity at various percentage resin, particle size of 0.045 mm and 30°C

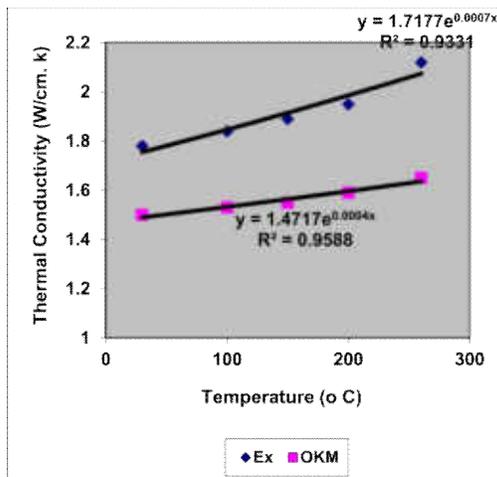
CONCLUSION

Based on the study the following concluding remarks can be given for the thermal conductivity of carbon resin electrodes:

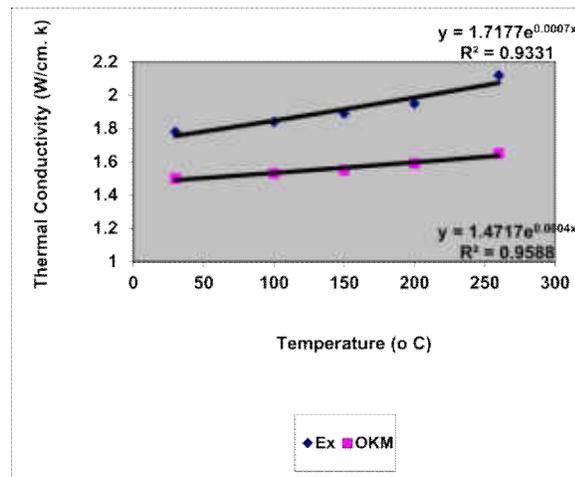
- Thermal conductivity of carbon resin electrode was less than thermal conductivity of heat treatment carbon (2.24–2.45 W/k.cm)
- Compacting pressure, heating temperature, percentage resin and

particle size of carbon play an important role on the thermal conductivity of carbon resin electrode,

- Increasing carbonization temperature and compacting pressure increases the thermal conductivity, and at higher carbonization of greater than 270°C carbon resin electrodes turned into ashes, and
- Thermal conductivity of the electrode



(a)



(b)

Figure 5: Relationship between experimental data and Okazaki et al model
 (a) comparison between experimental data and Okazaki et al. model
 (b) correlating experimental data with Okazaki et al. model

increases with decreasing percentage resin and particle size of carbon.

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